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Actinide and fission product partitioning and transmutation

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NUCLEAR ENERGY AGENCY
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ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

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The OECD Nuclear Energy Agency (NEA) was established on 1st February 1958 under the name of the OEEC European Nuclear Energy Agency. It received its present designation on 20th April 1972, when Japan became its first non-European full member. NEA membership today consists of 28 OECD member countries: Australia, Austria, Belgium, Canada, the Czech Republic, Denmark, Finland, France, Germany, Greece, Hungary, Iceland, Ireland, Italy, Japan, Luxembourg, Mexico, the Netherlands, Norway, Portugal, Republic of Korea, the Slovak Republic, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The Commission of the European Communities also takes part in the work of the Agency.

The mission of the NEA is:

- to assist its member countries in maintaining and further developing, through international co-operation, the scientific, technological and legal bases required for a safe, environmentally friendly and economical use of nuclear energy for peaceful purposes, as well as
- to provide authoritative assessments and to forge common understandings on key issues, as input to government decisions on nuclear energy policy and to broader OECD policy analyses in areas such as energy and sustainable development.

Specific areas of competence of the NEA include safety and regulation of nuclear activities, radioactive waste management, radiological protection, nuclear science, economic and technical analyses of the nuclear fuel cycle, nuclear law and liability, and public information.

The NEA Data Bank provides nuclear data and computer program services for participating countries. In these and related tasks, the NEA works in close collaboration with the International Atomic Energy Agency in Vienna, with which it has a Co-operation Agreement, as well as with other international organisations in the nuclear field.

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Foreword

Partitioning and transmutation (P&T) is one of the key technologies for reducing the radiotoxicity and volume of radioactive waste arisings. Recent developments indicate the need for embedding P&T strategies into advanced fuel cycles considering both waste management and economic issues. In this context, the OECD Nuclear Energy Agency (NEA) has been organising a series of biennial information exchange meetings on Actinide and Fission Product Partitioning and Transmutation to provide experts with a forum to present and to discuss developments in the field of P&T. The scope of the information exchange meetings covers all the major scientific topics related to P&T.

Previous meetings have been held in Mito (Japan) in 1990, at Argonne (USA) in 1992, in Cadarache (France) in 1994, in Mito (Japan) in 1996, in Mol (Belgium) in 1998, in Madrid (Spain) in 2000, in Jeju (Korea) in 2002, in Las Vegas (USA) in 2004 and in Nîmes (France) in 2006. The meetings have often been co-sponsored by the European Commission (EC) and the International Atomic Energy Agency (IAEA).

The 10th Information Exchange Meeting was held in Mito, Japan on 6-10 October 2008, comprising a special session on fuel cycle strategies and transition scenarios as well as six technical sessions covering various aspects of P&T. The meeting was hosted by the Japan Atomic Energy Agency.

The information exchange meetings on P&T form an integral part of NEA activities concerning advanced nuclear fuel cycles. An overview of NEA activities on P&T and relevant publications are available at *www.nea.fr/html/pt/welcome.html*.

These proceedings include all the papers presented at the 10th Information Exchange Meeting. The opinions expressed are those of the authors only and do not necessarily reflect the views of the NEA or its member countries.

Acknowledgements

The OECD Nuclear Energy Agency (NEA) gratefully acknowledges the Japan Atomic Energy Agency (JAEA) for hosting tenth Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation. The Agency also extends its gratitude to the European Commission (EC) and the International Atomic Energy Agency (IAEA) for their co-operation.

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Executive summary

Since 1990, the OECD Nuclear Energy Agency (NEA) has been organising, in response to the interest of member countries, a series of international information exchange meetings on the various aspects of partitioning and transmutation (P&T), providing experts with a forum to present and discuss current developments in the field.

Ten information exchange meetings have been organised thus far (Mito, Japan in 1990; ANL, USA in 1992; Cadarache, France in 1994; Mito, Japan in 1996; Mol, Belgium in 1998; Madrid, Spain in 2000; Jeju, Korea in 2002, Las Vegas, USA in 2004, Nîmes, France in 2006 and Mito, Japan in 2008). This tenth meeting was hosted by the Japan Atomic Energy Agency (JAEA) and was held in co-operation with the European Commission (EC) and the International Atomic Energy Agency (IAEA).

The meeting covered scientific, as well as strategic/policy developments in the field of P&T, such as: the role of P&T in advanced nuclear fuel cycles, including their impact on waste management policies; developments in partitioning and the managing the wastes produced; the technology of dedicated accelerator-driven systems as transmutation devices, including developments in accelerators, materials (targets and coolants) and fuels; the performance of advanced reactors, Generation IV and dedicated systems, for such application as transmutation as well as the safety of the latter; and R&D needs and results, including benchmarks, data improvements, experiments and the role of international collaboration. A total of 153 participants from 14 countries and 3 international organisations participated and delivered 117 presentations (45 oral presentations and 72 posters). A closing session with a panel discussion was also organised at the end of the meeting.

It was decided that progress reports from various countries and international organisations will be presented at alternate meetings. A specific subject is discussed during the meetings which do not call for the progress reports. This edition, a special session on fuel cycle strategies and transition scenarios was organised on the first day to discuss the different approaches and the tools, technology development, and R&D needed to support the transition from the current thermal to future fast reactors.

Opening session

Chairs: K. Minato, P. D'hondt

The opening session began with welcome addresses from Thierry Dujardin, NEA Deputy Director, Hideaki Yokomizo, Executive Director of the JAEA and Shunichi Tanaka, Vice Chairman of the Atomic Energy Commission of Japan. The welcoming speeches were followed by three presentations from the NEA, IAEA and the European Commission, covering current activities in these organisations.

- Th. Dujardin (NEA) gave an overview of ongoing NEA activities on actinide fission product partitioning and transmutation. Both scientific and strategic issues are horizontally considered in the framework of NEA programmes. A comprehensive handbook on lead and lead alloys and a report on the management of recyclable fissile and fertile materials were published. The topic of fuel cycle transition scenarios is being studied from the scientific and strategic points of view. Information exchange meetings on P&T and various other workshops are also being organised.

- P&T-related IAEA activities were presented by A. Stanculescu (IAEA). In the framework of the IAEA, P&T-specific projects are implemented in the nuclear power technology development and nuclear fuel cycle and material sections. Various activities of Technical Working Groups on Fast Reactors (TWG-FR) and on Nuclear Fuel Cycle Options (TWG-NFCO) were introduced. The IAEA organises conferences and schools in the field of advanced nuclear systems and maintains an accelerator-driven system R&D database. Future activities and goals were outlined.

- V. Bhatnagar (EC) summarised EU strategy in P&T and related programmes in the framework of EURATOM. The EU strategy as concerns P&T is based on sustainable use of nuclear energy. To implement P&T, various projects have been conducted within the FP6: EUROTRANS, EUROPART, RED-IMPACT, VELLA, etc. From 2007, FP7 organises new projects such as ACSEPT, GETMAT, F-BRIDGE, etc. The EU is pursuing cross-cutting collaboration between P&T and geological disposal.

One special session and six technical sessions, covering mainly scientific and technical issues in the field of P&T, were organised. Invited and selected papers were presented during these sessions. A poster session was also organised covering a variety of topics. The contents of the different sessions are described below, together with a brief explanation of the papers.

Special session: Fuel cycle strategies and transition scenarios

Chairs: M. Salvatores, K. Ono

This session comprised three invited papers and five oral presentations:

- K. McCarthy (INL, USA) was invited to present US activities on fuel cycle transition scenarios. US studies on the fuel cycle options are based on the guidance provided by systems analysis. Recent systems analysis activities are focused on system performance during transition to a closed fuel cycle. The study also covers economics of fuel cycles and transuranics (TRU) management.
- S. Kondo (JAEA, Japan) presented the current status of the Japanese nuclear fuel cycle. Based on the Japanese Fast Reactor (FR) cycle and the Fast Reactor Cycle Technology Development Project (FaCT), Japan is planning to deploy commercial FR around 2050. R&D activities on innovative reactor and fuel cycle technologies and Japanese transition scenarios were presented.
- F. Carré and J-M. Delbecq (CEA and EDF, France) gave a presentation on the French fuel cycle strategy and transition scenarios. Based on the national plan on the sustainable management of nuclear materials and a 2006 French law, France has planned to build a new prototype fast reactor in 2020. Transition scenarios regarding Gen-IV type reactors were also presented.
- M. Salvatores (CEA, France/INL, USA) presented the latest outcomes from the NEA activity on fuel cycle transition scenarios and the European approach. Taking into account current and future national planning with regard to nuclear strategy, four groups of countries and four possible transition scenarios were suggested and analysed. The results showed that regional strategies may provide a framework for implementing innovative nuclear fuel cycles. Future studies will cover fast reactor or accelerator-driven systems characteristics' optimisation and the impact P&T might have on geological disposal.
- K. Ono (JAEA, Japan) reported on the results of global scenarios for fast reactor deployment. The scenario is based on a homogeneous scenario employing a fast reactor with MOX fuel, which is currently being developed by Japan. Two representative scenarios were studied with assumptions concerning fuel, deployment time, breeding ratio, etc. Both scenarios were compared in terms of cumulative natural uranium demand, reprocessing capacity, nuclear power generation, etc.
- G. Dyck (AECL, Canada) discussed actinide transmutation in CANDU reactors, which may efficiently transmute TRU. Two transmutation scenarios were examined: group-extracted TRU in MOX and separated Am/Cm in targets. The results showed a significant increase in geological capacity. The full core calculations indicate that both fuel cycles are feasible.
- Materials assessments, which could be used in advanced nuclear fuel cycles from a safeguard perspective, were presented by C. Bathke (LANL, USA). The study mainly focused on UREX, COEX, THOREX and PYROX reprocessing to examine the attractiveness of materials mixture containing special nuclear materials (SNM). The conclusion indicated that there is a safeguard and security benefit with respect to diluting the reprocessing end products with lanthanides and uranium, and a related study on all possible means of reprocessing is needed.

Two additional papers were presented during the poster session.

Technical session I: Impact on P&T on waste management and geological disposal

Chairs: V.P. Bhatnagar, M. Nutt

This session comprised one invited paper and two oral presentations:

- H. Oigawa (JAEA, Japan) presented on the concept of waste management and geological disposal incorporating P&T technology. One of greatest benefits of P&T with regard to HLW management is to reduce size of the repository. The study indicates that the recovery and transmutation of MA can play an important role in stabilising the repository area for future Pu utilisation. When both UO_2 and MOX are used, the P&T process may reduce the conventional disposal area by up to five times.
- E.M. Golzalez (CIEMAT, Spain) gave a summary of the RED-IMPACT study, which is the study of the impact of P&T on the HLW management programme of the EC. The RED-IMPACT programme has been conducted by 24 laboratories from 12 EU countries. Six work programmes were organised to estimate the effect of P&T on waste management using existing deep geological repository designs. Scenarios for the advanced fuel cycle and human intrusion scenarios were also introduced.
- J-O. Liljenzin (Chalmers University, Sweden) presented an estimation of maximum permissible step losses in P&T processing. The calculation method of permissible step losses for continuous or batch-wise P&T operations from given values of the total acceptable loss fraction found in radioactive waste were introduced. In case of Np, and based on a relative hazard calculation, it was found that the acceptable step losses are much smaller in any continuous process than in a discontinuous process. The equations derived permit to compare the requirements on both the "P" and the "T" part of any P&T process for reduction of the amount of long-lived nuclides in a repository by a given factor.

Three additional papers were presented during the poster session.

Technical session II: Progress in transmutation fuels and targets

Chairs: D. Warin, Y. Arai

This session comprised one invited paper and seven oral presentations:

- The invited talk of the session was given by J. Somers (ITU, EC), on advanced fuel fabrication processes for transmutation. Since 2004, ITU has been working on various projects to develop transmutation fuels and related technologies. The presentation covered the SUPERFACT programme, infiltrations of HELIOS fuels, CERMET fuel development, nitride/carbide fuel fabrication technologies as well as future plans for Gen-IV fast reactors and ADS fuel development.
- K. Pasamehmetoglu (INL, USA) presented the development status of transuranic-bearing metal fuels in the USA. From the early 1990s, the USA has been developing metal fuels for sodium fast reactors. Metal fuel development for transmutation applications continues, including measurement of thermodynamic properties, phase diagrams/phase distribution/ microstructure, diffusion-couple measurements and fuel cladding development such as high-fluence data on HT-9 (200 dpa), ODS and lined/coated cladding options.
- F. Delage (CEA, France) summarised European projects on design, development and qualification of advanced fuels for an industrial ADS prototype. Oxide-based fuels are the reference model for ADS fuel in Europe and the nitride fuels are considered as the back-up solution. Results of the fuel design and performance assessment, safety analysis and in- and out-of-pile experiments are presented. Fuel thermochemical behaviour modelling is also under development.
- The Japanese study of the microstructural evolution and Am migration behaviour in Am-containing MOX fuels at the initial stage of irradiation was presented by K. Tanaka (JAEA, Japan). The JAEA is developing Am-MOX fuel pins containing 5% or 3% of Am. Ceramography results showed that structural changes occurred rapidly during irradiation. Fuel integrity was

maintained during the test. The conclusion indicates that careful consideration must be given to the redistribution behaviour of Am as well as to that of Pu for evaluating the impact on the thermal performance of Am-MOX fuels.

- Y. Arai (JAEA, Japan) discussed Japanese status on the development of nitride fuel and a pyrochemical process for transmutation of minor actinides. The nitride fuel cycle has been developed and its technical feasibility has been demonstrated. Studies regarding thermal properties of the nitride fuels, measurement of minor actinide nitride composite and related pyrochemical processes were summarised. Further study will include the preparation of thermal and thermodynamic database on minor actinide nitride fuel cycle.
- M. Kormilitsyn (RIAR, Russian Federation) presented the RIAR DOVITA-1/2 P&T programme, discussing the results of 15 years of R&D activity. RIAR has been studying pyrochemical processes since the 1960s. In 1992 RIAR initiated the DOVITA programme, which is dedicated to minor actinide fuel development through a pyrochemical process. The DOVITA-2 programme, initiated in 2006, covers minor actinide recycling in a fast reactor closed fuel cycle. Details of the programme, research results and future plans were presented.
- M. Kato (JAEA, Japan) presented an experimental evaluation of Am- and Np-bearing mixed-oxide fuel properties. The measurement results of physical properties, lattice parameters, phase diagram, oxygen potentials, melting temperatures and thermal conductivities of Am and Np-bearing MOX were presented. The effects of minor actinide addition on the physical properties are found to be negligible. Minor actinide addition effects on fuel properties were equally limited. The basic data for conducting the irradiation tests of the MA-bearing MOX fuel were established.
- D. Warin (CEA, France), discussed minor actinide recycling in sodium fast reactor and the 2008 status of the Phénix experimental programme. The specificities of minor-actinide-bearing fuels for recycling in SFR, the ongoing transmutation experiments in Phénix, post-irradiation examination at Phénix and the Phénix end-of-life test programme were presented. It was also announced that France recently established a programme to build a prototype Gen-IV SFR in 2020.

Seven additional papers were presented during the poster session.

Technical session III: Progress in partitioning, waste forms and management

Chairs: C. Hill, T. Inoue

This session comprised two invited paper and eight oral presentations:

- The first invited talk of the session, given by D. Warin (CEA, France) concerned future nuclear fuel cycles and the prospects and challenges lying therein. France has demonstrated a minor actinide partitioning strategy using the PUREX process with DIAMEX and SANEX to extract and separate minor actinides. To be in compliance with a 2006 French law, research programmes must demonstrate the industrial potentialities of the diverse P&T options by 2012. Advanced processes such as COEX, GANEX, PYRO, etc., as well as other future programmes, were presented.
- S. Bourg (CEA, France) gave an overview on a new EC activity: Actinide Recycling by Separation and Transmutation (ACSEPT), which is under the FP7 of EURATOM. Participants include 34 laboratories from 12 European countries and 4 non-EU countries with the objective to prepare future pilot-scale demonstrations of fuel treatment and refabrication of homo- and heterogeneous recycling of the minor actinides by either hydro- or pyro-metallurgy. Organisation of the programme, current activities and the work plan were presented.
- G. Modolo (FZJ, Germany) presented developments regarding a new SANEX process for actinide (III)/lanthanide (III) separation using a mixture of $CyMe_4BTBP$ and TODGA as selective extractants. The status of EUROPART research on An (III)/Ln(III) separation was summarised. The results showed that $CyMe_4BTBP$ is a promising extractant for An(III)/Ln(III) and the kinetic of extraction is low, and can be improved by a phase transfer catalyst such as DMDOHEMA or TODGA. Perspectives for further development were also summarised.

- Recent R&D activities pertaining to innovative extractants and adsorbents for partitioning of minor actinides at JAEA were reported on by T. Kimura (JAEA, Japan). Solvent extraction and extraction chromatography for the selective separation of An(III) have been investigated for the partitioning of minor actinides at JAEA. The study continues with the goal to implement separation technology at the industrial scale, especially for the separation of An(III) from RE.
- M. Nutt (ANL, USA) gave the second invited talk of the session, on fission product partitioning and integrated waste management. The partitioning of fission products in an advanced nuclear fuel cycle presents opportunities for improving the management of nuclear wastes. A systematic approach is needed to develop the entire waste management system, considering a broad suite of aspects. Activities are under way in GNEP/AFCI to develop/characterise waste forms and to optimise the waste management system through the implementation of an Integrated Waste Management Strategy.
- Studies on separation of actinides and lanthanides by extraction chromatography using 2.6-bis triazinyl pyridine were presented by K.N. Sabharwal (IGCAR, India). The second stage of the Indian nuclear power programme is to implement fast breed reactors. Various projects on fast reactor fuel development were addressed. The results of experimental work concerning separation chemistry were also summarised.
- T. Koyama (CRIEPI, Japan) presented recent developments on pyrochemical processing and metal fuel cycle technology at CRIEPI. Concerning fast breeder reactor fuel cycle technology, an engineering model of process equipments will be developed based on the detailed material balance obtained by hot examination. As for P&T study, tests of pyro-partitioning of actinides from real HLLW will be finished, and electrorefining of irradiated MA-bearing metal fuel will be carried out.
- E-H. Kim (KAERI, Korea) reported on fission product partitioning and waste salt minimisation during pyroprocess. Four types of waste stream have been considered: volatile and semi-volatile species waste stream; metallic form waste stream; uranium metal waste stream; and salt waste stream-ceramic composite waste form. The strategy of KAERI on waste minimisation focuses on the reduction of HLW through conversion to LLW and reduction of LLW which arises from voloxidation. Studies on the comparison of waste form volume with and without a salt recycle as well as a demonstration plan for the engineering scale were presented.
- Current progress in R&D on MSR fuel cycle technology in the Czech Republic was presented by J. Uhlíř (NRI, Czech Republic). A successful solution for MSR/MSTR fuel cycle technologies represents an essential precondition for the future deployment of MSR systems. Fluoride pyrochemical separation methods seem to be promising technologies for use within these fuel cycles. Current R&D efforts and results achieved thus far offer the prospect that MSR/MSTR fuel processing and reprocessing will be successfully resolved.
- J-H. Ahn (UC Berkeley, USA) presented the effects of repository conditions on environmental impact reduction through recycling. The study aims to compare the environmental impacts of a water-saturated repository and the Yucca mountain repository coupled with fuel cycles. The results indicate that without separation of TRU, the level of the impact mainly depends on the repository condition and solidification matrix and for the Yucca mountain repository, the effect of separation efficiencies appear proportionally on the environment impact.

Thirty-four additional papers were presented during the poster session.

Technical session IV: Progress in materials, including spallation targets and coolants

Chairs: C. Fazio, S. Monti

This session comprised one invited paper and two oral presentations:

- The topic of the invited talk of the session, presented by C. Fazio (FZK, Germany) was progress in materials, including spallation targets and coolants. Background and a discussion of past works was provided, as was news concerning FP6 and European projects on P&T materials such as DEMETRA (EUROTRANS), the VELLA and ELSY projects and their latest results. For the

FP7, the GETMAT project which covers Gen-IV material development was initiated in 2008. A view on the MEGAPIE experiment beyond the materials assessment was also presented. MEGAPIE aims to demonstrate the feasibility of a liquid metal target for high-power spallation ADS applications. Post-test results on thermo-hydraulics and neutronics as well as prospective ADS application of the test data were presented.

- K. Kikuchi (JAEA, Japan) presented progress on materials development in a lead-bismuth (LBE) spallation target system. Many test programmes have been performed at the Transmutation Experimental Facility (TEF), an ADS target test facility of the Japanese ADS project, J-PARC. The goal of the test is the acquisition of LBE control techniques. Test results of LBE loops of corrosion-erosion, target model and thermal fluid were summarised.
- I-S. Hwang (SNU, Korea) reported on corrosion tests of FeCr and FeCral alloys in static and flowing LBE. Based on test and model, the total oxide thickness of FMS steels was predicted at 450°C. Corrosion behaviours of alumina-forming alloys and conventional FeCr alloys have been investigated. The formation of Cr sulphide appears to promote void formations at the oxide/alloy interface, weakening the interfacial bonding and leading to severe dissolution attack in LBE. A wall-thinning model for the magnetite layer and means for the suppression of segregation in Al-containing alloys are to be studied in the future.

Two additional papers were presented during the poster session.

Technical session V: Progress in transmutation physics experiments and nuclear data

Chairs: H. Oigawa, E.M. Gonzalez Romero

This session comprised one invited paper and three oral presentations:

- The invited talk of the session concerned nuclear data for advanced fuel cycles and was presented by M. Salvatores (CEA, France). The goal of the study was to perform a systematic approach to define data needs for advanced reactors and the fuel cycle. The study has been performed by NSC Working Party on Evaluation Co-operation (WPEC). A sensitivity and uncertainty study has given the impact of neutron cross-section uncertainty on the integral parameters of the core and fuel cycle of a wide range of innovative systems that correspond to advanced nuclear systems. Main findings and results were presented.
- D. Cano Ott (CIEMAT, Spain) presented the cross-section measurements of minor actinides from the n_TOF-Ph2 experiment at CERN, Switzerland. The study aims to improve differential neutron cross-sections for minor actinides. n_TOF is a unique facility for high performance detectors for fission (PPAC and FIC) and capture (TAC) cross-section measurements, with a fully digital data acquisition system. This technology has already been proven useful for Np and Pu. The n_TOF operation was interrupted in 2004 and will be restarted in November 2008.
- T. Sasa (JAEA, Japan) gave an overview on the status of J-PARC's Transmutation Experimental Facility (TEF). J-PARC started construction in March 2002, and the spallation neutron was successfully produced in May 2008. The TEF aims to study ADS development, minor actinide management, nuclear data, shielding safety, LBE target and medical applications. The results of recent studies and planned future projects were also presented.
- M. Fernández-Ordóñez (CIEMAT, Spain) reported on the reactivity monitoring of the YALINA subcritical assembly using beam trips and current-to-power experiments. The YALINA facility is to qualify the reactivity monitoring techniques as well as to develop the electronic chains that can be used in a power ADS. The experimental set-up, PNS experiments, current-to-power experiment and beam trip experiments and their results were summarised.

Six additional papers were presented during the poster session.

Technical session VI: Transmutation systems: design, performance and safety

Chairs: I-S. Hwang, K. McCarthy

This session comprised one invited paper and five oral presentations:

- The invited talk of the session concerned trends in transmutation performance and safety parameters versus TRU conversion ratio of sodium-cooled fast reactors and was presented by W-S. Yang (ANL, USA). The United States' GNEP fast reactor campaign and advanced burner reactor plan were presented. For the parametric study, core design and oxide and metal fuel core configurations were explained. The results show that the TRU consumption rate reaches ~80% of the maximum theoretical value when the TRU conversion ratio is in the range 0.25-0.35. As the TRU conversion ratio decreases, the initial heavy metal inventory and charge rate decrease, however, the TRU inventory and charge rate slightly increases. The estimated safety parameters indicate that the metal and oxide cores of the TRU conversion ratio in the range of 0.25-0.40 are acceptable for passive safety features.
- C. Artioli (ENEA, Italy) discussed the effectiveness of the ELSY concept with respect to minor actinide transmutation capabilities. The ELSY and adiabatic core configuration for the ELSY lead fast reactor were presented. The result of the core demonstration, minor actinide concentration at equilibrium state, adiabatic core cycle and its mass flow were summarised. Future work includes a demonstration using the Pu equilibrium vector in a system with a unitary breeding ratio.
- X-N. Chen (FZK, Germany) presented design, safety and fuel developments for the EFIT accelerator-driven system with CERCER and CERMET cores. The European Facility for Industrial Transmutation (EFIT) has been developed under the framework of FP6 of EUROTRANS project. The CERCER core has been chosen as the reference core and CERMET for the alternative core. The past and current status of both cores' development was presented. Fuel and material behaviour of both cores as well as future plans were also presented.
- A design study of minor-actinide-bearing oxide fuel core for homogeneous TRU recycling fast reactor system was presented by S. Ohki (JAEA, Japan). Within the framework of the fast reactor development project (FaCT), Japan is studying a conceptual design of a sodium-cooled MOX fuel core, considering TRU composition change during the reactor transition from LWR to FR. The conclusion showed that homogeneous TRU recycling may solve the problem of increasing fresh-fuel decay heat due to the source nuclides.
- J-W. Jang (KAERI, Korea) presented core performance and safety implications of TRU burning medium-to-large fast reactor core concepts. The study aims to investigate TRU burning capability in various sizes of fast reactors and to identify constraints of core scale-up studies. The results showed that the TRU burning rates are almost identical to the various core sizes in terms of performance and the reactivity coefficient of the system. Future work will cover conversion ratio changes and safety evaluation of the suggested core design.
- F. Varaine (CEA, France) summarised outcomes from the study of minor actinide transmutation in a sodium fast reactor depleted uranium radial blanket. Transmuting minor actinides in the radial blanket has the advantage of proliferation resistance. Two different cores were suggested and analysed. The results indicate that 20% of minor actinide in the blanket is acceptable. The fuel behaviour, fuel fabrication technologies, handling, transportation, etc. were identified for the needs of future R&D.

Eighteen additional papers were presented during the poster session.

Closing session

Chair: K. Minato

A closing session was held at which session Chairs were invited to present highlights from their technical sessions and to form a panel for an open discussion with the attendees.

It was agreed that the special session was successful and benefited from a well-chosen topic. Such a special session will be organised every two meetings. Fuel cycle strategies and transition scenarios are being studied world wide and by international organisations either in regional or global perspectives. P&T technologies embedded transition scenario studies as well as their impact on geological disposal should be further expanded upon for the successful deployment of advanced nuclear systems.

The technical sessions were discussed based on the four parameters proposed by K. Minato: main achievements; future needs; technical obstacles; recommendations. Progress was made in the area of minor-actinide-related science and technologies and uncertainty analysis of nuclear data. Many of the results were produced from international experimental projects. P&T-related system design studies have remarkably increased in quantity. For the successful implementation of industrial-scale P&T, the demonstration of related technologies and fuel cycle scenario studies integrating the partitioning process are needed. Stable political and financial support is indispensable and economics and cost benefit studies are necessary as well. However, the cost of P&T-related facilities is still too high and transportation issues are not yet resolved.

The Chairs recommended continuing strong international collaboration and developing cross-cutting programmes in the field of P&T. Communication and discussion with the geological disposal community with the common goal of radioactive waste management must be pursued. Maintaining a close relationship with industry and regulatory bodies will also facilitate the future deployment of the P&T-embedded fuel cycles.

K. McCarthy (INL, USA) announced INL's willingness to host the 11th Information Exchange Meeting in the United States. There was also a proposal from the Czech Republic to host the 12th Information Exchange Meeting in 2012.

The meeting was closed by K. Minato, the Chair of the Scientific Advisory Committee. Technical visits to JAEA sites were arranged for the following day.

Welcome

OECD/NEA welcome address

Thierry Dujardin
OECD Nuclear Energy Agency

On behalf of OECD Nuclear Energy Agency, it is a great honour for me to welcome you all to the 10th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation.

As you well know, Mito is an historical place for NEA P&T meetings, as the first and fourth meetings in this series of information exchange meetings were held here in 1990 and 1996 respectively. I would like to express my sincere appreciation to our hosts, the Japan Atomic Energy Agency and the meeting chair, Dr. Kazuo Minato, as well as all JAEA staff and the members of the scientific advisory committee, who have worked hard in preparing this meeting.

I would also like to thank the European Commission and the International Atomic Energy Agency, here represented by Drs. Ved Bhatnagar and Alexander Stanculescu, for their co-operation in the organisation of this meeting.

I am very pleased to see that the interest in partitioning and transmutation is still very high after almost 20 years of activity. NEA involvement in the field emerged from an initiative in 1988 by the Japanese government to launch a long-term research and development programme on the recycling and transmutation of actinides and long-lived fission products, the so-called OMEGA programme. The NEA was invited to conduct an international project related to actinide separation and use. The present series of information exchange meetings is one of the outcomes of this international project and provides an opportunity for scientists to present and discuss the results of their research with colleagues. This tenth meeting is the largest meeting ever in terms of number of papers, especially in the fields of partitioning, waste forms and management, as well as design, performance and safety of transmutation systems.

The two NEA committees, the Nuclear Science Committee and the Nuclear Development Committee, that organise this series of meetings, have introduced a novelty at this year’s meeting. They have decided that at every second meeting there will be a special session dedicated to a specific field of research, replacing the reports on national programmes. This year’s special session will be devoted to fuel cycle strategies and transition scenarios, a topic included in both NEA committee programmes of work that I will briefly develop in my presentation later this morning.

Recent years may be looked at as a turning point in the consideration of nuclear energy as a viable option for future sustainable energy policies. The main reasons for this nuclear comeback on the world energy agenda are well-known: rising fossil fuel prices and increasing concerns about climate change and the security of energy supply. However, there is still a gap between a comeback on the agenda and an effective and efficient role of nuclear energy in the future world energy mix. As the Generation IV International Forum stated, future nuclear energy systems will require better economics, increased safety and reliability, enhanced resistance to proliferation, improved physical protection, a better use of natural resources and a better management of waste. In many OECD countries the latter goal is the key issue to increase public acceptance.

With its potential to strongly reduce the amount and the radiotoxicity of nuclear waste ultimately disposed of, partitioning and transmutation and advanced fuel cycles offer opportunities which should be considered among future options to increase the public confidence in the safe and sustainable management of high-level radioactive waste. At the last NEA P&T meeting, held at Nîmes, France, in

September 2006, it was noted that optimisation of the fuel cycle through partitioning and transmutation must take into account waste management issues, for all waste streams within the fuel cycle, including their associated costs and risks.

I would like to also take this opportunity to inform you that the 50th anniversary of the NEA will be celebrated next week in connection with the meeting of the NEA Steering Committee. During the celebration session more than 300 dignitaries and government experts, including ministers from many countries, the OECD Secretary General and the IAEA Director General, will discuss the future of nuclear energy for peaceful purposes and challenges for new nuclear build. In connection with this anniversary the NEA will also publish a book entitled *Nuclear Energy Outlook*, a first of a kind for the NEA, aiming to respond to the renewed interest in nuclear energy by many OECD countries. It uses the most current data and statistics available and provides projections up to 2050 to consider growth scenarios and potential implications on the future use of nuclear energy. It also offers unique analysis and recommendations on the possible challenges that lie ahead.

Before closing my welcome address, I am happy to announce that the NEA has received a proposal from the Idaho National Laboratory in USA to host the next information exchange meeting on P&T in the second half of 2010. The details will be announced in near future.

Ladies and gentlemen, unfortunately, I will not be able to stay with you until Thursday, but looking at the programme, I am convinced that you will have a very interesting and fruitful meeting. I wish you all a very enjoyable stay in Mito.

JAEA welcome address

Hideaki Yokomizo
Japan Atomic Energy Agency

I am Hideaki Yokomizo, the executive director of Japan Atomic Energy Agency. On behalf of the JAEA, I would like to extend a cordial welcome to all of you, gathering here for this 10th OECD/NEA Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation.

Nowadays, nuclear power has become a very important energy resource as one of the solutions against the global environmental issues of the emission of greenhouse effect gases. As demand for nuclear energy grows, future nuclear systems consisting of advanced reactors, accelerator-driven systems and associated fuel cycles have been proposed and developed aiming at more effective use of nuclear fuels and reasonable waste disposal.

We believe that partitioning and transmutation is one of the most promising options for the enhancement of the efficiency of high-level waste disposal and the utilisation of resources in the spent fuel. Japan had already launched a long-term R&D plan on partitioning and transmutation technology in 1988, called the OMEGA programme, and has invited the NEA to co-ordinate an international co-operation programme for information exchange on this important issue.

It is a great pleasure and honour for us to have hosted the first NEA Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation in 1990, and that this 10th meeting is taking place here. We believe that this co-operation programme has benefited and enhanced the studies in this field and will be expanded onward.

In Japan, the Atomic Energy Commission has started the check and review partitioning and transmutation technology in the working group under the Advisory Committee on Research and Development. This working group will review the progress on Japanese partitioning and transmutation technologies since 2000, and will discuss the future R&D plan.

JAEA is now preparing to restart operation of Monju, a FBR-type reactor, which has not been in operation since 1995 due to a sodium leakage in its secondary heat transfer system during performance tests. Monju will be ready to operate early next year. As the number of fast reactors for research is decreasing in the world, we believe that the Monju will be an important R&D tool for transmutation system.

Also, Phase I of the J-PARC project is almost complete and about to start its service. In the second phase of J-PARC, the Transmutation Experimental Facility will be constructed, though the budget of the construction has not yet been approved. I also hope this Phase II project becomes an international project and that the JAEA plays an important role in the R&D of partitioning and transmutation technology in the world.

I sincerely hope that this 10th meeting will be fruitful and useful for all the participants and provides opportunities to establish further international collaborations. Also, I would like to express my deepest gratitude to Mr. Dujardin of the Nuclear Energy Agency (NEA), the members of the Scientific Advisory Committee, and all the participants.

In closing my remarks I wish you an enjoyable stay. Autumn is the best season in Japan. I hope you will be refreshed with the nice foods and atmosphere of Mito as well as the animated discussion at the meeting. Thank you very much for your attention.

AEC welcome address

Shunichi Tanaka
Atomic Energy Commission of Japan

Good morning ladies and gentlemen, it is a great honor to have a chance to address you at this 10th OECD Nuclear Energy Agency Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation.

On behalf of the Atomic Energy Commission, I would like to commend the OECD/NEA for its excellent leadership initiative as regards the meeting organisation and promotion of the policy and research on partitioning and transmutation. Also, I am very delighted that Japan is once again hosting this meeting, as it did in 1990 and 1996.

Nuclear policy of Japan

First let me introduce briefly the nuclear policy of Japan. In 2005, the Atomic Energy Commission decided upon a framework for nuclear energy policy, in which we categorised important actions into the three classifications of short, medium and long term.

The short-term actions are those aiming at using existing power plants as long as practicable, plants for which the life expectancy is to be extended more than 50 to 60 years. These actions include activities for maintaining public confidence in the safety management of existing plants and related facilities, improving the performance of existing plants and promoting the use of Pu (plutonium) recovered from the spent fuels in LWR. In particular, one of the most crucial issues in the short term is to make progress in site finding for geological disposal of high-level wastes.

One of the actions in the mid-term is to prepare advanced and innovative LWR for replacement of retiring plants, as a number of existing plants will be retired in 10 to 30 years.

Long-term actions are those related to the development of innovative nuclear energy systems which can be competitive with other technology in the future market in terms of social acceptability as well as safety, economy and environmental protection. The R&D in this domain includes nuclear science and technology research on fusion reactor and partitioning and transmutation (P&T) with accelerator-driven system (ADS). And, one of the major projects in this group of actions is the development of a fast breeder reactor and its fuel cycle system, which is expected to be commercially available in the 2050s.

Management of HLW

Nuclear energy is one of the most effective means to both ensure a steady energy supply and to reduce global emission of greenhouse gases. It is equally necessary to ensure the safety, security and non-proliferation of nuclear energy.

At the same time, the management of high-level radioactive waste is one of the most crucial issues limiting the expansion of nuclear energy. The policy in Japan is to dispose vitrified wastes in deep underground below more than 300 m. No disposal site, however, has been decided as of today, as public acceptance and trust on its safety has been difficult to acquire. In this regard, the situation may be more or less common in every country. But this issue is more immediate for Japan, because vitrified high-level wastes will soon be produced by the Rokkasho commercial reprocessing plant, which will soon begin full-scale operation.

Role of P&T

Now, let's turn to P&T. I believe that there are two roles for P&T:

- One is to reduce public concern for geological disposal of HLW with very long-life nuclides as MA. This idea is a rather classical reason of MA transmutation.
- The other is a little bit special, while more essential. Now, it is planned in Japan that 40 000 to 50 000 vitrified HLW be stored at one disposal site. While, 55 nuclear power plants are in operation in Japan, supplying about 50 MWe. As a result, about 1 000 spent fuel assemblies are produced every year, and processed into almost same number of vitrified HLW in a reprocessing plant. The situation means that a new disposal site will be needed every 40 to 50 years, if the present policy is kept. This hurdle is too high to overcome for us.

You know that the disposal site can be used efficiently by more than 4 to 5 times, if long-life exothermic FP such as ^{137}Cs, ^{90}Sr and MA are removed from HLW. And, needless to say, it will bring invaluable benefit in disposal policy and cost, as well. These are the reasons why we have promoted the R&D for P&T as the long-term action.

Assessment of P&T by AEC

The research and development of P&T has been carried out under international and domestic frameworks for almost 20 years. The AEC recently initiated an assessment of P&T, where the technical feasibility of transmutation is discussed, focusing on transmutation with FBR and ADS, and partitioning technology of MA and FP. The assessment is very important for determining the future of P&T policy in Japan.

The AEC seeks innovative FBR cycle technologies with such performances as safety, reliability, commercial competence, high breeding ratio for fuel utilisation, nuclear proliferation resistance, and mitigation of environmental burden. FBR has, in physics, a potential for MA transmutation with its hard fast neutron spectrum, while it is very important to figure out for us whether the transmutation of MA is compatible with other criteria required by a nuclear technology system or not. For example, the additional increase of Na void coefficient in a MA core might significantly affect the critical safety design. The breeding performance is essential to fast reactor, and to what extent is it influenced by MA burning? It is also pointed out that the manufacture and treatment of MA fuel assemblies is very difficult in commercial base because of large nuclear heat emission and strong radiations.

I would like to ask all participants to help clarify these issues. Because we have an idea that the deployment plan of FBR in market place must not be delayed by the requirement of MA transmutation. Essential performances of FBR are breeding, safety, reliability and commercial competence. I have concern that the requirements of AEC for next FBR system may be too stringent in this regard.

For ADS system, it does not seem that sufficient and essential information has been classified for subcritical MA burning system with high-power accelerator as of yet. I think that the R&D of ADS still remains in the fundamental stages. I would like to ask you to prepare an R&D road map for the ADS system based on current knowledge under international co-operation. Right now, we are confronted with a difficulty, whether R&D facilities should be constructed or not in J-PARC. The judgement should be made considering the scientific discussion of international community. In this regard, a reliable road map is essential for our decision.

Further, we must pay more attention to partitioning, as well. It is very important to make clear what kinds of nuclides have to be separated for P&T, and to assess the state of the art of partitioning technology, since the partitioning system definitely influences the future process of spent fuels, and even fuel cycle including next-generation reprocessing plant.

Closing

The AEC-Japan is going to determine a policy of P&T covering management of spent fuels in the near future. In this context, this meeting is very timely, and I am sure that it will provide an invaluable opportunity for sharing experiences, exchanging information and generating ideas for international co-operation. Let me wish you all success in this meeting.

Thank you for your kind attention.

Opening session

Chairs: K. Minato, P.J. D’hondt

Overview of NEA activities in actinide and fission product partitioning and transmutation

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Abstract

Since 1989, the OECD Nuclear Energy Agency (NEA) has been conducting international studies related to partitioning and transmutation (P&T) of nuclear waste, covering issues related to the physics and chemistry of P&T, including accelerators, targets, and fuel and structural materials, as well as fuel cycle and radioactive waste management strategies. The information exchange meetings on Actinide and Fission Product Partitioning and Transmutation, organised under the auspices of the Nuclear Development Committee (NDC) and the Nuclear Science Committee (NSC), are part of this programme. Their objective is to enhance the value of basic research in the field of P&T by providing experts with a forum to present and discuss current developments in the field and by supporting international collaborations. This paper provides an overview of results from recent NEA activities in the field of P&T and gives an insight into ongoing projects and planned future activities.

Introduction

The OECD/NEA focuses mainly on P&T-related scientific and strategic issues. The activities involving both issues provide a good example of the NEA's horizontal activity. Most of the work on P&T is carried out under the auspices of the Nuclear Science Committee (NSC) and the Nuclear Development Committee (NDC). The NSC's work in this field is mainly focused on scientific aspects of P&T, whereas the work of the NDC mainly consists of strategic and assessment reports on the broad field of P&T and its impact on the economics of the nuclear fuel cycle. Recently, fuel cycle transition scenario activity has been studied from both a scientific and a strategic perspective.

Organising the Information Exchange Meetings on Actinide and Fission Product Partitioning and Transmutation has been an important activity in the P&T area. The meetings were initiated in 1989 following a proposal from the Japanese government requesting that an international information exchange programme on issues related to P&T be set up [1]. Since the first meeting, which was held in Mito, Japan in 1990, the scope of the meetings has expanded from basic P&T technology to transmutation system design. Now, the potential implementation and industrialisation of P&T are under discussion. The meetings have been held every two years in different NEA member countries. The forthcoming 11th meeting, hosted by Idaho National Laboratory (INL), will be held in San Francisco, USA, in November 2010. The NEA will also organise two other P&T related workshops in 2010, one on Technology and Components of the Accelerator-driven Systems (TCADS) in Germany and one on Structure Materials for Innovative Nuclear Systems (SMINS) in Korea.

Scientific issues

Handbook on Lead-bismuth Eutectic

Heavy liquid metal (HLM) cooled systems are one among several options for future nuclear power plants. The cooling materials (lead and lead-bismuth eutectic alloys) are chemically inert and their boiling temperature is very high. In addition, they have good natural circulation characteristics. In 2007, the NSC published a comprehensive handbook on the properties of lead-bismuth eutectic alloys [2]. The purpose of the handbook is to develop standards, identify areas where further studies are needed, and help establish a common methodology for experiments and data analyses. Eight institutes and national laboratories from seven member countries contributed to the study. The structure of this handbook is as follows: four chapters are dedicated to heavy liquid metal properties; four chapters cover materials and testing issues; two chapters summarise key aspects of the thermal-hydraulics, instrumentation and system technologies. The last three chapters present information on existing test facilities and safety guidelines. An updated version of the handbook is planned to be published in 2010, including material test results from the Russian Federation and a best practice guide, based on the heavy liquid metal coolant loop thermal-hydraulics study.

Fuel cycle flow sheet studies

A critical analysis of the partitioning system and separation technologies based on separation performance, as well as the quantity and nature of waste and the cost of processing need to be carried out to properly evaluate various P&T scenarios. The NSC is analysing flow sheets of the various processes used in the reprocessing of nuclear fuel in order to evaluate critical performance parameters for each process and scenario. It is expected that the detailed mass balance flow sheet will characterise high-level waste generation and assess technical needs. Expected technical performances, such as recovery efficiency, product decontamination, waste radiotoxicity and decay heat generation, etc., are evaluated as well. The flow sheet of aqueous, pyrochemical and fluoride processes are being studied. These studies are based on coated-particle, oxide, metallic and nitride fuels. A total of 12 flow sheets are being collected from eight countries including the Russian Federation. The report will be published in 2010. The following processes are covered:

- standard PUREX;
- UREX+3 (Pu/Np recovery, minor actinide separation);
- grind/leach;

- pyrochemical (non-fertile nitride);
- two-step pyrochemical (reduction to metal, followed by electrochemical steps);
- pyrochemical (direct electrochemical processing of oxide);
- extended PUREX (Pu/Np recovery, minor actinide separation);
- fluoride volatility;
- pyrochemical (direct electrochemical processing of metallic fuel);
- pyrochemical (FR nitride);
- advanced PUREX (innovative features).

New activities in preparation

The following new activities have been initiated under the auspices of the NSC.

Minor actinide bearing fuels for advanced nuclear systems will be studied with the objective to identify technical issues and to support development of innovative fuels, including cladding materials, by assessing state-of-the-art innovative fuel development and by establishing an experimental database. Benchmark studies on fuel performance codes and experiments will also be performed.

The separation and management of curium, which dominates the heat generation and radiotoxicity of spent nuclear fuel, will be studied. A report will be prepared, covering the status of national research on curium, as well as advantages, possible options and recommendations on how to manage curium.

The NSC will also make a comparative study on homogeneous vs. heterogeneous recycle of transuranics (TRU) in fast reactors and review the potential benefits and impacts of advanced fuel cycles with P&T. The kick-off meetings of these activities will be held in the first half of 2009.

Strategic issues

Management of recyclable fissile and fertile materials

Interest in nuclear energy continues to grow in many countries as a means of providing a more secure energy supply and reducing greenhouse gas emissions from the power sector. In this context, recyclable materials constitute an asset for broadening the resource base for nuclear fuel supply, especially from a medium to long-term standpoint [3]. The NDC has studied this question and has issued a report providing an overview of recyclable fissile and fertile materials inventories, which can be re-used as nuclear fuel. The report describes the options available for managing those materials, through recycling and/or disposal. The potential energetic value of recyclable materials is being assessed while taking into account the variability of retrievable energy contents in various materials according to technology and strategy choices made by those who own the materials. The analyses contained in this report will be of particular interest to energy policy makers and to nuclear fuel cycle experts. The study's conclusions highlight the importance of recyclable fissile and fertile materials in view of future nuclear energy development, taking into account sustainable development goals. Furthermore, the analyses show that technical options are available or under development for management, by means of recycling and/or disposal, of all material inventories.

Status report on national programmes in partitioning

The NSC is preparing a state-of-the-art report on national programmes in partitioning, covering both aqueous and pyrochemical processes. Information on the national P&T research programmes in eight member countries, as well as the Russian Federation and the European Commission, are being collected. The objectives are to perform a thorough technical assessment of separation processes as applied to a broad set of P&T scenarios, identifying important research, development and demonstration issues necessary to bring preferred technologies to a deployable stage and to recommend collaborative international efforts to further technology development. The publication of the report is planned for the first half of 2009.

Fuel cycle transition scenarios

Transition from current fuel cycles to long-term more sustainable fuel cycles is one of most important items on the agenda for 21st century nuclear R&D. To meet the demand from member countries, the NEA is conducting various projects in the field of fuel cycle transition scenarios from both scientific and strategic perspectives. The NSC is ready to publish a state-of-the-art report that provides a framework for assessing national needs regarding fuel cycle transitions and covers the timing of key technologies.

The NSC is also conducting a benchmark study that compares the performance of various codes, performing depletion and transition scenario calculations. Another study, based on a European region scenario, was recently finalised. This study considers the implementation of innovative fuel cycles associated with partitioning and transmutation in Europe. The study shows the impact of different deployment strategies and of different country policies on the suggested regional transition scenarios. Regional facility characteristics and potential deployment schedules are also discussed. The report will be published during the first half of 2009. A scenario covering the projections of world-wide energy demand, nuclear energy share and deployment of advanced nuclear systems, accounting for different world regional characteristics, is also being studied.

The NDC has studied the transition from thermal to fast nuclear energy systems from a strategic and policy view, covering national, regional and global issues. The report notes that although fast neutron systems are attractive at the global level for enhancing security of nuclear fuel supply and reducing the volumes and radiotoxicity of waste, their relevance and viability in a given country highly depends on each national and regional context. In particular, the size of the national nuclear fleet and its expected growth rate are key factors in assessing the interest of introducing fast neutron systems. The radioactive waste management policy of the country also determines the relevance of fast neutron systems as an option for facilitating waste management and disposal.

The NDC study also notes that for countries with a small fleet of reactors in operation and with modest expected growth of nuclear electricity generation, building the infrastructure to support the transition from thermal to fast neutron reactor systems is unlikely to be cost effective, but could nevertheless be considered in a broad sustainable development perspective. In such countries, multilateral agreements and international co-operation may be required to ensure the viability of reprocessing and recycling spent nuclear fuel in fast neutron reactors.

The need to evaluate the advantages and drawbacks of transition scenarios in a holistic approach is also highlighted taking into consideration short-term and long-term aspects, and assessing environmental and social criteria as well as economics. Its conclusions show that the viability of transition scenarios and their successful implementation will require long-term commitments and comprehensive and consistent planning.

Meetings and workshops

Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation

Since 1990, the NEA has organised a series of information exchange meetings for discussion of scientific and strategic developments in the field of P&T. The 10th meeting attracted a total of 120 abstracts from 14 countries and three international organisations. The abstracts covered the following topics:

- impact of P&T on waste management and geological disposal;
- progress in transmutation fuels and targets;
- progress in partitioning, waste forms and management;
- progress in materials, including spallation targets and coolants;
- progress in transmutation physics experiments and nuclear data;
- transmutation systems design, performance and safety.

A special session on fuel cycle strategies and transition scenarios was also organised. Compared to the 9th meeting in 2006, the number of papers on materials, spallation targets and coolant technology decreased from 37 to five. However, papers on transmutation systems have increased from 16 to 24.

As with the 9th meeting, the majority of papers were in the field of partitioning technology, waste forms and management (42 papers in 2006 and 43 papers in 2008). The 11th Information Exchange Meeting will be held in November 2010 in San Francisco, USA and will be hosted by INL. The Czech Republic's NRI has offered to host the 12th meeting in 2012.

Workshop on Technology and Component of ADS (TCADS)

Since 1998 the NEA has been organising workshops on the "Utilisation and Reliability of High Power Proton Accelerators (HPPA)" to discuss issues of common interest and to present the most recent achievements in the areas of accelerator reliability, which is critical to building accelerator-driven systems. The 5th workshop (HPPA5) was held on 6-9 May 2007 in Mol, Belgium and was hosted by SCK•CEN [4]. A total of 42 papers was presented. They covered topics such as targets, windows and coolant technologies, subcritical system design and ADS simulations, safety and control of ADS as well as ADS experiments and test facilities. A conclusion from the HPPA5 meeting was that the reliability of high power proton accelerators has improved. As a consequence, the workshop scope has been extended to cover not only accelerator, but also subcritical system design and component development. Agreement was, therefore, reached to change the title of the workshop from "Utilisation and Reliability of High Power Proton Accelerators (HPPA)" to "Technology and Component of ADS (TCADS)" in order to meet member countries' demands. The first TCADS workshop will be held during the first half of 2010 in Germany and will be hosted by FZK.

Workshop on the Structural Materials for Innovative Nuclear Systems (SMINS)

Following a proposal from Germany to the NSC, a Workshop on the Structural Materials for Innovative Nuclear Systems was held on 4-6 June 2007 in Karlsruhe, Germany and was hosted by FZK [5]. The workshop was attended by approximately 100 scientists and, following five keynote speeches, the following four technical sessions were covered:

- materials for very high temperature reactors;
- materials for metal-cooled reactors;
- material for water-cooled reactors;
- multi-scale modelling.

Organisation of the second SMINS workshop has begun.

Concluding remarks

The OECD Nuclear Energy Agency (NEA) carries out a wide range of activities in the field of partitioning and transmutation in response to the needs of its member countries. These activities deal with scientific, as well as strategic, issues and their large number illustrates that reducing the amount of high-level waste over the long term is an important criterion for NEA member countries. In planning its future activities in the area of P&T, the NEA is relying on continued feedback from its member countries to correctly prioritise its activities in this field.

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IAEA activities in the area of partitioning and transmutation

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Abstract

The major challenges facing the long-term development of nuclear energy as a part of the world's energy mix are improvement of the economic competitiveness, meeting increasingly stringent safety requirements, adhering to the criteria of sustainable development, and public acceptability. The concern linked to the long life of many of the radioisotopes generated from fission has led to increased R&D efforts to develop a technology aimed at reducing the amount of long-lived radioactive waste through transmutation in fission reactors or accelerator-driven hybrids. In recent years, in various countries and at an international level, more and more studies have been carried out on advanced and innovative waste management strategies (i.e. actinide separation and elimination). Within the framework of the Project on Technology Advances in Fast Reactors and Accelerator-driven Systems (www.iaea.org/inisnkm/nkm/aws/fnss/index.html), the IAEA initiated a number of activities on utilisation of plutonium and transmutation of long-lived radioactive waste, accelerator-driven systems, thorium fuel options, innovative nuclear reactors and fuel cycles, non-conventional nuclear energy systems and fusion/fission hybrids. These activities are implemented under the guidance and with the support of the IAEA Nuclear Energy Department's Technical Working Group on Fast Reactors (TWG-FR). The paper presents an overview of these activities.

Introduction

Based on an experience of more than 105 reactor-years, nuclear power is a mature technology that makes a large contribution to the energy supply world wide. As of August 2008, there were 439 nuclear power plants operating in the world with a total net installed electrical capacity of 372 GW supplying slightly more than 15% of the world's electricity, and 35 nuclear power plants under construction [1]. In 2007, the nuclear share in electricity generation ranged from maxima of 76.9%, 64.4% and 54.3% in France, Lithuania and Slovakia, respectively, to minima of 2.8%, 2.5%, 2.3% and 1.9% in Brazil, India, Pakistan and China, respectively [1].

According to the projections published by the Intergovernmental Panel on Climate Change (IPCC), the median electricity increase until 2050 will be by a factor of almost 5. It is reasonable to assume that nuclear energy will play a role in meeting this demand growth. However, there are four major challenges facing the long-term development of nuclear energy as a part of the world's energy mix: improvement of the economic competitiveness, meeting increasingly stringent safety requirements, adhering to the criteria of sustainable development and public acceptability. Meeting the sustainability criteria is the driving force behind the topic of this paper. More specifically, in this context sustainability has two aspects: natural resources and waste management. IAEA's activities in the area of partitioning and transmutation (P&T) are mostly in response to the latter. While not involving the large quantities of gaseous products and toxic solid wastes associated with fossil fuels, radioactive waste disposal is today's dominant public acceptance issue. In fact, small waste quantities permit a rigorous confinement strategy, and mined geological disposal is the strategy followed by some countries. Nevertheless, political opposition arguing that this does not yet constitute a safe disposal technology has largely stalled these efforts. One of the primary reasons that are cited is the long life of many of the radioisotopes generated from fission. This concern has led to increased R&D efforts to develop a technology aimed at reducing the amount of long-lived radioactive waste through transmutation in fission reactors or accelerator-driven hybrids. In recent years, in various countries and at an international level, more and more studies have been carried out on advanced and innovative waste management strategies (*i.e.* actinide separation and elimination). Within the framework of the Project on Technology Advances in Fast Reactors and Accelerator-driven Systems (*www.iaea.org/inisnkm/nkm/aws/fnss/index.html*), the IAEA initiated a number of activities on utilisation of plutonium and transmutation of long-lived radioactive waste, accelerator-driven systems, thorium fuel options, innovative nuclear reactors and fuel cycles, non-conventional nuclear energy systems, and fusion/fission hybrids. The paper presents an overview of these activities.

IAEA activities

As in all the other fields of advanced nuclear power technology development, the IAEA is relying also in the P&T area on broad, in-depth staff experience and perspective. The framework for all the IAEA activities in the P&T area is the Technical Working Group on Fast Reactors (TWG-FR). In responding to strong common R&D needs in the member states, the TWG-FR acts as a catalyst for international information exchange and collaborative R&D.

Given the common technical ground between plutonium utilisation R&D activities and the development of technologies for the transmutation and utilisation of long-lived fission products and actinides, both activities are performed within the framework of a single IAEA project: Technology Advances in Fast Reactors and Accelerator-driven Systems [2].

The TWG-FR is a standing working group within the framework of the IAEA. It provides a forum for exchange of non-commercial scientific and technical information, and a forum for international co-operation on generic research and development programmes on advances in fast reactors and fast spectrum accelerator-driven systems. Its present members are the following 14 IAEA member states: Belarus, Brazil, China, France, Germany, India, Italy, Japan, Kazakhstan, Republic of Korea, Russian Federation, Switzerland, United Kingdom and the United States of America, as well as the ISTC, OECD/NEA and the EU (EC). The TWG-FR has also welcomed, as observers, Belgium and Sweden. The TWG-FR advises the Deputy Director General-Nuclear Energy on status of and recent results achieved in the national technology development programmes relevant to the TWG-FR's scope, and recommends activities that are beneficial to these national programmes. It furthermore assists in the implementation of corresponding IAEA activities, and ensures that through continuous consultations with officially nominated participants in the annual co-ordination meetings of the TWG-FR all the project's technical activities performed within the framework of the Nuclear Power Technology Development sub-programme are in line with expressed member state needs. The scope of the TWG-FR is broad, covering all technical aspects of fast reactors and ADS research and development, design, deployment, operation and decommissioning. It includes, in particular: design and technologies for current and advanced fast reactors and ADS; economics, performance and safety of fast reactors and ADS; associated advanced fuel cycles and fuel options for the utilisation and transmutation of actinides and long-lived fission products, including the utilisation of thorium. Given the TWG-FR's broad scope, the coverage will generally be in an integrative sense to ensure that all key technology areas are covered. Many specific technologies are addressed in detail by other projects within the IAEA and in other international organisations. The TWG-FR keeps abreast of such work, avoiding unproductive overlap, and engages in co-operative activities with other projects where appropriate. The TWG-FR thus co-ordinates its activities in interfacing areas with other Agency projects, especially those of the International Working Group on Nuclear Fuel Cycle Options, and the Department of Nuclear Safety, as well as with related activities of other international organisations (ISTC, OECD/NEA and EC).

Recent and ongoing activities

Three education and training activities were implemented by the IAEA in collaboration with the International Centre for Theoretical Physics (ICTP):

- School on *Physics, Technology and Applications of Accelerator-driven Systems (ADS)* [3]. The purpose of the school was to familiarise students with the status of the R&D activities in the ADS area. A review of the ADS designs presently under consideration was given. The participants studied the theoretical foundations of all ADS design aspects (*i.e.* high-power accelerator, spallation target and sub-critical core) and identified the most problematic areas, as well as the limitations of the simulation methods presently used. The students familiarised themselves with the modern theoretical models used to predict nuclear reaction cross-sections. They studied the principles of the evaluation methodology and became acquainted with the existing data libraries and the data processing and transport calculations. Based on the discussion of the impact of the present uncertainties on ADS performance, further needs for data and methods development and validation work were be identified.
- Advanced Workshop on *Model Codes for Spallation Reactions* [4]. The major objective of the workshop was to improve the understanding of the physical basis, approximations, strengths and weaknesses of the currently used spallation codes. Specifically, the workshop promoted in-depth understanding of the physics of INC, QMD models and de-excitation models. It pointed out reasons for the respective successes or deficiencies of different models and helped to define an agreed set of experimental data to be used in validation and inter-comparison of the models. The presentation of relevant basic experimental data with emphasis on accuracies, detector efficiencies, filters and thresholds created the basis for such benchmarking and inter-comparison activities. The participants agreed to organise, under the auspices of IAEA, an international benchmark exercise of the different models. The specifications of the benchmark, including the set of selected experimental data, were defined during the workshop.
- Workshop on *Nuclear Reaction Data for Advanced Reactor Technologies* [5]. The workshop provided training and information exchange opportunities to nuclear physicists, nuclear engineers and other users of nuclear data for advanced technological applications. Starting from the facilities and techniques used for the experimental determination of basic nuclear data, and continuing with the evaluation procedures and creation of reliable nuclear reaction data libraries, the lectures summarised the most recent developments and results in this field. The following topical areas were addressed: facilities and instrumentation for nuclear reaction data measurements; experimental techniques for measurements of nuclear reaction data; modern theoretical nuclear structure and reaction models; evaluation of nuclear data and creation of evaluated nuclear reaction data libraries; simulation tools for design studies of advanced nuclear systems; and overview of strategies for advanced nuclear technologies.

With regard to collaborative R&D, the IAEA has completed the Co-ordinated Research Project (CRP) on Studies of Advanced Reactor Technology Options for Effective Incineration of Radioactive Waste, and has an ongoing CRP (2005-2009) on Analytical and Experimental Benchmark Analyses of Accelerator-driven Systems (ADS).

The CRP on Studies of Advanced Reactor Technology Options for Effective Incineration of Radioactive Waste concentrated on the assessment of the dynamic behaviour of various transmutation systems. The reactor systems investigated were categorised in eight domains and comprise critical reactors, subcritical accelerator-driven systems with heavy liquid metal and gas cooling, critical molten salt systems and hybrid fusion/fission systems. For all reactor systems fertile and fertile-free fuel options have been investigated. A major effort of the CRP consisted in the benchmarking of steady-state core configurations and performing transient/accident simulations.

For a general assessment and comparison, the relevant safety coefficients were determined for the individual systems. In a second step transient analyses were performed which reflected the generic behaviour of the various reactors types. Issues as the transmutation potential, burn-up behaviour and decay heat of minor-actinide-bearing fuels were also investigated.

The transmutation systems with a high minor actinide load generally show deteriorated safety parameters. Benchmarking of calculational tools and data is therefore mandatory.

The results of the CRP show that for steady-state analyses the neutronic tools in each domain are advanced enough to provide good agreement. This holds for both mechanistic SN and Monte Carlo codes. Larger spreads in the results are generally caused by the different nuclear data libraries used. These deviations may not only be caused by the minor actinide data but also by data of other constituents, *e.g.* the treatment of matrix material in inert fuels and the fission products. Transient calculations have been performed for all transmutation systems except the gas-cooled ADS. Very different code systems were employed from point-kinetics to space-time kinetics, and also different levels in the sophistication of the thermal-hydraulics modelling. The benchmarking leads to the conclusion that different level code systems are currently needed to cover all the time scales of the different systems and transients. The very detailed codes have difficulties in their running times, for instance in the case of a long-lasting loss of heat sink accidents, while the less detailed codes naturally neglect important phenomena. A need for an intermediate class of codes becomes obvious. With one exception (ADS with fertile-free fuel), the benchmarking has exclusively been performed in the range of transients and accidents without core disruption.

The comparison of the dynamic behaviour of the different systems allowed identifying the intrinsic transient behaviour and time scales of these systems, as well as the dominating feedback effects. For fertile systems the prompt Doppler feedback is the important balancing effect. In non-fertile systems this part is taken over by the subcriticality or by the structural thermal expansion. Also noteworthy is the slow and sluggish dynamic behaviour of the molten salt systems, compared to the other systems investigated. An important observation to be made is that the comparison of the dynamic behaviour of the different systems has to be balanced in the sense that, while a large knowledge base exists *e.g.* for the critical fast reactors, much less is known for other systems like fusion-fission hybrid systems. Nevertheless, characteristic transients, phenomena and time scales were already identified by the CRP. The final report of the CRP has been drafted and will be published in 2009.

The objective of the CRP on Analytical and Experimental Benchmark Analyses of Accelerator-driven Systems (ADS) is to improve the understanding of the physics of the coupling of external neutron sources with subcritical cores. The CRP participants are performing computational and experimental benchmarking for ADS and non-spallation neutron source driven subcritical systems. In the first stage, the CRP comprises the following work domains [6,7]: YALINA Booster; Kyoto University Critical Assembly (KUCA); Pre-TRADE; FEAT and TARC; ADS kinetics analytical benchmarks; actinide cross-sections; spallation targets; and ADS performance. So far, the progress achieved by the CRP is not uniform in all the work domains. In some cases, intermediate results reveal high discrepancies, requiring further in-depth analyses.

Last but not least, the IAEA has implemented the ADS Research and Development Database. It provides information about ADS-related R&D programmes, existing and planned experimental facilities as well as programmes, methods and data development efforts, design studies and so forth. While operational on the Internet and open to all users (*http://www-adsdb.iaea.org/index.cfm*), the database has to rely on content contributed by the interested community. Data and information can be provided online, and contributions are solicited (the author will gladly provide, upon request, access privileges as editor to anyone wishing to contribute content).

Planned activities

In collaboration with ICTP, IAEA's Department of Nuclear Energy and Department of Nuclear Sciences and Applications are organising the School on Physics, Technology and Applications of Innovative Fast Neutron Systems, in Trieste, Italy, from 9-20 November 2009. The syllabus of the school includes Physics and Design Studies; General Concept Studies; Sub-system Studies; Nuclear Data; Fuel Development; Fuel Recycling; Fuel Cycle Studies; and the Impact of Transmutation Scenarios on High-level Waste Repositories.

Two major international conferences addressing topics relevant to this paper are being organised by the IAEA in 2009: the International Topical Meeting on Nuclear Research Applications and Utilisation of Accelerators, organised in collaboration with the ANS by IAEA's Department of Nuclear Energy and Department of Nuclear Sciences and Applications, in Vienna, 4-8 May 2009 [8]; and the conference on Fast Reactors and Associated Fuel Cycle – Challenges and Opportunities, organised by IAEA's Department of Nuclear Energy[1], and hosted by JAEA in Kyoto from 7-11 December 2009 [9].

Two major status report publications relevant to the topics of this paper are planned for 2009, viz. the *Accelerator-driven Systems: Energy Generation and Transmutation of Nuclear Waste; Status Report*, and the *Status Report on Lead and Lead-bismuth-cooled Fast Reactors*.

Conclusions

Renewed interest in nuclear energy is driven by the need to develop carbon-free energy sources, demographics and development in emerging economies, and security of supply concerns. The pace at which the nuclear energy option is embraced is accelerating world wide, with the existing marked imbalances in energy availability causing more and more emerging economies to give it serious consideration. For obvious sustainability reasons, spent fuel utilisation and breeding are returning to centre stage, and with this the fast reactor as the necessary linchpin. The necessary condition for successful deployment of fast neutron systems in the near and mid-term is the understanding and assessment of technological and design options, based on both past knowledge and experience, as well as on research and technology development efforts. With regard to the latter, achieving the full potential of the fast neutron system and closed fuel cycle technologies with regard to both efficient utilisation of the fissile resources and waste management is conditional on continued advances in research and technology development to ensure improved economics and maintain high safety levels with increased simplification of fast neutron systems. The utilisation of breeding to secure long-term fuel supply remains the ultimate goal of these systems. Nevertheless, plutonium recycling in fast reactors, as well as incineration/transmutation of minor actinides and long-lived fission products in various hybrid reactor systems (*e.g.* ADS) offers promising waste management options. Several R&D programmes in various member states are actively pursuing these options, along with the energy production and breeding mission of fast reactor systems. In line with the statutory objective expressed in Article II (*The Agency shall seek to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world. It shall ensure, insofar as it is able, that assistance provided by it or at its request or under its supervision or control is not used in such a way as to further any military purpose.*), the IAEA will continue to assist the member states' activities, also in the area of advanced technology development for utilisation and transmutation of actinides and long-lived fission products, by providing an umbrella for information exchange and collaborative R&D to pool resources and expertise.

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1. In co-operation with the Japan Atomic Energy Commission Ministry of Economy, Trade and Industry (Japan), Ministry of Education, Culture, Sports, Science and Technology (Japan), Japan Atomic Industrial Forum, Inc., Wakasa Wan Energy Research Centre (Japan), American Nuclear Society, Atomic Energy Society of Japan, European Nuclear Society, Institute of Electrical Engineers of Japan, Japan Society for Mechanical Engineers, Korean Nuclear Society, European Commission and the OECD Nuclear Energy Agency.

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[9] IAEA, Meetings Web Page,
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List of related IAEA publications

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EU strategy in partitioning and transmutation and its implementation within the Euratom Framework Programmes

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Abstract

A robust strategy is followed in the European Union (EU) in the area of partitioning and transmutation (P&T) for sustainability of nuclear energy by promoting collaborative research and training among member states within the Euratom multi-annual Framework Programmes (FP) and Sustainable Nuclear Energy Technology Platform (SNE-TP). Once-through cycle ("disposal of spent fuel as is") does not appear to be sustainable for nuclear energy production. Reprocessing of the spent fuel and transmutation of minor actinides in dedicated devices would reduce considerably the radiotoxic inventory of the disposed waste in geological repositories. This is of significant importance in non-proliferation strategy and radiological terrorism and reduces risks in case of an inadvertent human intrusion. The separation of main heavy metals (uranium and plutonium) reduces the volume and thermal output of the waste to be disposed of, which increases effectively the capacity of the repository. Furthermore, extraction of heat-bearing components (Sr and Cs) from the waste "can reduce thermal output" of the disposed waste. Concerns of the public related to long-life of the waste could largely be overcome by P&T as it would reduce half-life of most of the waste to be disposed of to a couple of hundred years and it could thus come to the aid of geological disposal community in securing a broadly agreed political solution of waste disposal in geological repositories. Nevertheless, additional cost, additional secondary waste, activation products, intermediate-level waste and dose to workers in the process of P&T itself will contribute to defining an optimal transmutation scheme. A double-strata approach with subcritical accelerator-driven systems (ADS) and/or critical fast reactors (Generation IV systems) is being considered. A decision on the choice is planned in a couple of years. Geological disposal of the remaining waste (separation/transmutation losses) will nevertheless be necessary. Reprocessing of the spent fuel would be required no matter what path of transmutation is followed. Therefore efforts for the advanced partitioning processes are being reinforced towards pilot and test facilities for optimised separation processes in close co-operation with fuel fabrication teams and geological disposal community. The efforts and resources allocated to P&T have an added value of training many researchers contributing to the retaining and building of competence in the nuclear fission area. The above strategy is being implemented through a well balanced portfolio of Euratom Framework Programme research projects in the area of P&T.

1 Introduction

Promotion of collaborative research and training in nuclear energy in member states of the European Union (EU) has been at the heart of the Euratom Treaty (1957) and, since its inception, it has acted as a driving force for the development of nuclear power in the EU. For improving its competitiveness and the long-term sustainable economic growth, the EU has to secure its energy supply, reduce its dependency on imported energy, diversify its energy sources and develop new technologies taking into account the environmental challenges that it faces.

European Community research has been organised in Framework Programmes (FP) of durations of four to five years since 1984. In this regard, one of the objectives of EU is to achieve greater co-operation between member states' research strategies and a mutual opening-up of programmes. With the challenges and prospects opened up by the technologies of the future, there is a need to more thoroughly integrate European research efforts and capacities. To this end, the European Commission launched the so-called European Research Area (ERA) initiative in January 2000 [1]. Framework Programme research activities are geared toward making ERA a reality.

The current Framework Programme is FP7 (2007-2011). The priorities for the European Union's research and development activities for the period 2007-2011 are set out in the 7th Euratom Framework Programme (FP7) [2]. The FP7 focuses on research areas such as nuclear waste management including geological disposal and P&T, safety of existing reactor systems and advanced Gen-IV reactor concepts, radiation protection, support for research infrastructures and retaining competences and know-how in all areas of nuclear science. The FP6 [3] focused on a number of research areas combining technological, industrial, economic and social aspects.

P&T aims to ease the nuclear waste management problem by coming to the aid of geological disposal community in securing a broadly agreed political solution of waste disposal in geological repositories. P&T also endeavours to contribute to the sustainability of nuclear energy. Euratom Framework Programme (currently FP7 and the preceding FP6) research projects in the area of P&T are key elements in contributing to the implementation of this strategy.

2 EU strategy in the area of partitioning and transmutation

Sustainability of nuclear energy requires that future electricity producing nuclear systems are integrated into a closed fuel-cycle. Future Generation IV fast-reactor systems have the potential to produce 50 to 100 times more electricity than current reactors for the consumption of the same amount of uranium [4]. They are expected to produce significantly lower quantities of waste. P&T is expected to strongly minimise the waste legacy for geological disposal as compared to present reactors in terms of volume, thermal load and radiotoxic inventory.

A double-strata approach utilising energy producing reactors in conjunction with dedicated subcritical accelerator-driven systems (ADS) and/or critical fast reactors (Gen-IV systems) for waste transmutation has strong merits and has a good chance of achieving success. This scheme compartmentalises the tasks of electricity production and radioactive waste transmutation leading to an efficient implementation of both tasks fairly independently. It permits the utility to concentrate on electricity production without jeopardising its operation by unnecessarily further complicating its task by requiring it to transmute the waste in the same reactor. This additional task has a strong influence on the safety of energy producing critical fast reactor as well as it runs the risk of delays with the regulatory approval. The latter is highly constrained with the public safety and any innovation has to be systematically and safely demonstrated in a step-wise manner before the regulatory approval can be acquired. A dedicated waste transmutation system using an ADS and/or critical FR is therefore being considered. A decision on the choice is planned in a couple of years. Note that the geological disposal of the remaining waste (separation/transmutation losses) will nevertheless be necessary.

Once-through cycle ("disposal of spent fuel as is") does not appear to be sustainable for nuclear energy. Reprocessing of the spent fuel and transmutation of minor actinides in dedicated devices would reduce considerably the radiotoxic inventory of the disposed waste in geological repositories. This is of significant importance in the strategy of non-proliferation together with the mitigation of the danger of exposure in the unlikely event of a "human intrusion scenario" whereby a borehole is inadvertently drilled right through the repository. The separation of main heavy metals (uranium and plutonium) reduces the volume and thermal output of the waste to be disposed of, which effectively increases the capacity of the repository. Furthermore, an extraction of heat-bearing components (Sr and Cs) from the waste can reduce the thermal output of the disposed waste. This again permits a reduction in the size of the repository, except possibly in salt media which does not need this separation. Safe surface storage of these components (containing the long-lived ^{135}Cs) needs to be confirmed [5].

Concerns of the public related to long life of the waste could largely be overcome by P&T as it would reduce the half-life of most of the waste to be disposed of to a couple of hundred years and it could thus come to the aid of geological disposal community in securing a broadly agreed political solution of waste disposal in geological repositories. Nevertheless, additional cost, additional secondary waste, activation products, intermediate-level waste and dose to workers in the process of P&T itself will contribute to defining an optimal transmutation scheme.

Generation IV safe, advanced nuclear reactor concepts that burn waste and produce fuel for further use are gaining increased attention as a possible future course of action. Scientific and technical debate is ongoing, and it is not clear whether this path will or will not be followed and whether they will replace partially or fully the subcritical accelerator-driven system (ADS) activities for transmutation that are presently being pursued in the EU. Nevertheless, it is clear that reprocessing of the spent fuel would be required no matter what path of transmutation is followed. Therefore efforts for the advanced partitioning processes are being reinforced towards pilot and test facilities for optimised separation processes in close co-operation with fuel fabrication teams and geological disposal community. The latter should especially take account of the requirements and accommodate the waste streams emanating from the advanced (minor actinide) reprocessing systems with a view to transmutation whether in subcritical or critical devices.

The efforts and resources that are presently being allocated to the study of P&T have an added value of training many researchers by supporting a rather large number of PhD students through integrated projects. This will definitely contribute to the retaining and building of competence in the nuclear fission area. Moreover, even if ADS systems are found to be inappropriate for future research in waste transmutation, the efforts devoted to fuel, technology and nuclear data research will to a large extent be applicable to lead-cooled fast critical systems.

3 The Euratom 6th Framework Programme (FP6) (2002-2006)

The scientific and technical goal of the Euratom FP6 specific programme “Research and Training Programme on Nuclear Energy” [6] is to help exploit the full potential of nuclear energy, both in the long and short term. Its development and exploitation is to be done in a sustainable manner while combating climate change and reducing the energy dependency of the EU. Research and development activities in this programme have been subdivided into: *a)* fusion energy research; *b)* management of radioactive waste; *c)* radiation protection; *d)* other activities in the field of nuclear technologies and safety.

In area *b)*, the priority is to find a permanent and safe solution for the management of long-lived, high-level waste that is acceptable to society. This includes establishing a sound technical basis for the demonstration of long-lived high-level waste disposal in geological formations. This is to be supported by studies on P&T and further supplemented by exploring the potential of system concepts that would by themselves produce less waste in nuclear energy generation. Combating the decline in both student numbers and teaching establishments by a better integration of European education and training in nuclear safety and radiation protection is another important aim.

The detailed work programme of Euratom FP6 was adopted by the EC in December 2002 [7] and has been updated regularly. In P&T, the research areas include a fundamental assessment of the system and safety aspects of the overall concept of P&T and, in particular, of its impact on waste management and geological disposal. In the area of partitioning, continued R&D of hydrometallurgical and pyro-chemical processes is envisaged with a view to a demonstration of the most promising techniques. In the area of transmutation, the development of basic knowledge and technologies for transmutation and evaluation of their industrial practicability, in particular, of transmutation devices such as accelerator-driven subcritical systems (ADS) is proposed.

4 The research activities on P&T in the Euratom 6th Framework Programme

The projects in the area of P&T that were selected for funding in Euratom FP6 are given in Table 1. The reader is referred to each project's respective website for details of the objectives and results achieved and current status (see Table 2). The PATEROS project is a two-year Co-ordination Action establishing a global P&T roadmap leading up to the industrial scale deployment of necessary facilities. A common objective of all strategies using P&T is to reduce the burden on a long-term waste management, in terms of radiotoxicity, volume and heat load of high-level nuclear waste which has to be disposed of in final repositories. Possible strategies can range from using dedicated transmuters in a separate fuel cycle stratum in a stable or expanding nuclear energy scenario in order to reduce drastically the amount of nuclear waste to be sent to the repository, down to the scenario of a nuclear phase-out. RED-IMPACT is a three-year project studying the impact of P&T, conditioning and waste reduction technologies on reducing the burden associated with radioactive waste management and disposal. P&T is likely to ease the final repository requirements and it will also contribute to the sustainability of nuclear energy in those countries that pursue this source of energy. EUROPART is a three-year project studying the development of methods for the separation of individual minor actinides that are contained in aqueous nuclear wastes issuing from the reprocessing of uranium oxide (UOX) or mixed-oxide (MOX) nuclear spent fuel. It also studies separation of grouped actinides (An) for recycling. Partitioning techniques used are: *i)* hydrometallurgy; *ii)* pyro-metallurgy. The flowsheet of various processes including the conditioning methods for the wastes to be generated by the partitioning processes are also established. Processes for possible industrialisation of partitioning strategies have also been looked at.

Table 1: Euratom FP6 P&T projects underway indicating their salient features

SN	Acronym	Abbreviated title	Budget (M€)		Co-ordinator	Start date and duration
			Total	EC		
1	PATEROS	P&T European roadmap	0.8	0.6	SCK•CEN (BE)	01/09/2006 24 months
2	RED-IMPACT	Impact study of P&T on waste management	3.9	2.0	KTH (SE)	01/03/2004 36 months
3	EUROPART	Partitioning techniques and processes	11.2	6.0	CEA (FR)	01/01/2004 42 months
4	EUROTRANS	All aspects of transmutation by subcritical ADS	45.0	23.0	FZK (DE)	01/04/2005 60 months
5	ELSY	Waste transmutation in lead-cooled critical system	6.9	2.95	ANSALDO (IT)	01/09/2006 36 months
6	PUMA	Pu and MA management by thermal gas-cooled system	3.7	1.85	NRG (NL)	01/09/2006 36 months
7	VELLA	Networking of lead loop infrastructures in Europe	3.3	2.3	ENEA (IT)	01/10/2006 36 months
8	LWR-DEPUTY	LWR fuels for deep burning of Pu in thermal systems	2.4	1.25	SCK•CEN (BE)	01/08/2006 48 months
9	EFNUDAT	Networking of EU facilities for nuclear data measurements	3.0	2.4	CNRS (FR)	01/11/2006 48 months
10	CANDIDE	Networking of nuclear data for EU industrial development	0.8	0.8	UU (SE)	01/01/2007 24 months
11	NUDAME	Trans-national access for nuclear data measurements	0.2	0.2	EC-JRC (IRMM)	01/04/2005 36 months
		Totals	**81.0**	**43.5**		

EUROTRANS is a five-year project carrying out a preliminary detailed design of a ≈100 MW experimental facility demonstrating the technical feasibility of transmutation in an accelerator-driven system (XT-ADS) in the short term as well as to accomplish a reference conceptual design (several 100 MW) of a modular generic European Transmutation Demonstrator (ETD) over the long term. A zero-power lead fast critical facility connected to a continuous beam neutron generator is also being operated to allow a validation of the on-line subcriticality monitoring, the validation of neutronic calculation codes to reduce design safety margins and to support licensing applications for the construction of an experimental device. It also carries out studies on fuels, materials and neutron data for relevant isotopes. ELSY is a three-year project studying lead-cooled fast critical systems with a view to demonstrating that it is possible to design a competitive and safe system using simple engineered technical features. Safe burning of recycled minor actinides in the core of ELSY will also be studied. PUMA is a three-year project dealing with important issues concerning the use of Pu and MA in gas-cooled reactors and to provide additional key elements for the utilisation and transmutation of Pu and MA in current and future (high-temperature) gas-cooled reactor designs, contributing to the reduction of Pu and MA stockpiles, and to the development of safe and sustainable reactors. VELLA is a three-year Integrated Infrastructure Initiative project. It aims to create a virtual European laboratory for lead technologies. Its final goal is the creation of a network of the EU laboratories that operate devices using heavy liquid metal technologies, especially lead alloys. LWR-Deputy is four-year project conceived to fit into a portfolio of experimental research projects on novel fuels for deep burning of plutonium in existing nuclear power plants (NPP). It will investigate to what extent the existing NPP in Europe can create markedly less nuclear waste by moving to inert matrix fuels.

EFNUDAT is a four-year project planning to integrate all infrastructure-related aspects of nuclear data measurements by organising networking activities to optimise the use of the facilities for nuclear data measurements and the analysis and dissemination of results, Transnational Access Activities procuring approximately 4 000 additional beam hours for external users that will carry out nuclear data measurements, and Joint Research Activities to raise the performance of the facilities and the efficiency of their use. CANDIDE is a two-year co-ordination action to establish a durable networking of nuclear data efforts that are important in the context of minimising the high-level waste stream of nuclear energy. The purpose is to identify the needs for improved nuclear data, assess the present status of knowledge, and to estimate what accuracy can be reached with state-of-the-art techniques for the relevant fast critical reactors and subcritical ADS. NUDAME is three-year project aiming to promote transnational access to facilities at IRMM-JRC, Geel for neutron data measurements.

Table 2: Euratom FP6 P&T projects websites

SN	Acronym	Website addresses
1	PATEROS	www.sckcen.be/pateros/
2	RED-IMPACT	www.red-impact.proj.kth.se/
3	EUROPART	www.europart-project.org/scripts/home/publigen/content/templates/show.asp?L=EN&P=55
4	EUROTRANS	http://nuklear-server.ka.fzk.de/eurotrans/
5	ELSY	www.elsy-lead.com/
6	PUMA	www.puma-project.eu/
7	VELLA	www.3i-vella.eu
8	LWR-DEPUTY	www.sckcen.be/lwrdeputy/
9	EFNUDAT	http://nuclear-astrophysics.fzk.de/efnudat/
10	CANDIDE	http://candide.nri.cz/
11	NUDAME	www.irmm.jrc.be/

5 The Euratom 7th Framework Programme (FP7) (2007-2011)

Research and development activities in FP7 [8] comprise two themes: *i)* fusion energy; *ii)* nuclear fission and radiation protection. Research and development activities in the latter theme include: *a)* management of radioactive waste, *b)* reactor systems, *c)* radiation protection and cross-cutting activities; *d)* infrastructures; *e)* human resources, mobility and training. The objective of activities with regard to the management of radioactive waste are: to support the implementation-oriented research and development activities on all remaining key aspects of deep geological disposal of spent fuel and long-lived radioactive waste and, as appropriate, demonstration of the technologies and safety, and to underpin the development of a common European view on the main issues related to the management and disposal of waste. Research pertaining to reactor systems includes the study of partitioning and transmutation and/or other concepts aimed at reducing the amount and/or hazard of the waste for disposal. In the cross-cutting activities, the objectives of infrastructures are to support the availability of, and co-operation between, research infrastructures such as material test facilities, underground research laboratories and radiobiology facilities and tissue banks, necessary to maintain high standards of technical achievement, innovation and safety in the European nuclear sector. The human resources, mobility and training endeavour to support the retention and further development of scientific competence and human capacity (for instance through joint training activities) in order to guarantee the availability of suitably qualified researchers, engineers and employees in the nuclear sector over the longer term.

Moreover, RTD in all technical areas of partitioning and transmutation (P&T) will be supported in Euratom FP7, which could be the basis for the development of pilot facilities and demonstration systems for the most advanced partitioning processes and transmutation systems, involving subcritical and critical systems, with a view to reducing the volumes and hazard of high-level long-lived radioactive waste issuing from treatment of spent nuclear fuel. Research will also explore the potential of concepts that produce less waste in nuclear energy generation, including the more efficient use of fissile material in existing reactors.

Calls for proposals for the Euratom FP7 are made at a yearly interval. First and second calls of Euratom FP7 were made on 22 December 2006 and 30 November 2007 respectively. A number of proposals in the area of P&T have been funded (see next section) and the projects are well underway. The third call is scheduled to be published on 19 November 2008 with a deadline for the receipt of proposals of 21 April 2009.

6 The research activities on P&T in the Euratom 7th Framework Programme

The projects in the area of P&T that were selected for funding in Euratom FP7 are displayed in Table 3. The reader is referred to each project's respective website for details of the objectives and results achieved and current status (see Table 4).

Table 3: Euratom FP7 P&T projects underway indicating their salient features

SN	Acronym	Title	Budget (M€) Total	Budget (M€) EC	Co-ordinator	Start date/ end date
1	ACSEPT	Actinide recycling by separation and transmutation	23.8	9.0	CEA (FR)	01/04/2008 31/03/2012
2	GETMAT	Gen-IV and transmutation materials	14.0	7.5	FZK (DE)	01/03/2008 29/02/2013
3	EUFRAT	European facility for innovative reactor and transmutation neutron data	0.5	0.5	JRC-IRMM (Mol-BE)	01/10/2008 30/09/2012
4	ACTINET-I3	ACTINET integrated infrastructure initiative	~6.0	~3.0	CEA (FR)	Under negotiation
5	FAIRFUELS	Fabrication, irradiation and reprocessing of fuels and targets for transmutation	~6.5	~3.0	NRG (NL)	Under negotiation
6	CDT	Central design team for a fast spectrum transmutation experimental facility	~5.0	~2.0	SCK•CEN (BE)	Under negotiation
		Totals	**~55.8**	**~25.0**		

Table 4: Euratom FP7 P&T projects websites

SN	Acronym	Website addresses
1	ACSEPT	www.acsept.org
2	GETMAT	http://nuklear-server.ka.fzk.de/getmat/index.php
3	EUFRAT	http://irmm.jrc.ec.europa.eu/html/activities/eufrat/index.htm
4	ACTINET-I3	Under construction
5	FAIRFUELS	Under construction
6	CDT	Under construction

A four-year project on partitioning (ACSEPT) was started on 1 April 2008 after the project EUROPART (FP6) was successfully completed. ACSEPT is a structured R&D framework to develop chemical separation processes compatible with fuel fabrication techniques, with a view to their future demonstration at the pilot level. The project plans to optimise and select the most promising processes dedicated to actinide partitioning and those featuring a group separation. An exploratory research focused on the design of new molecules will also be conducted. Moreover, pyroprocesses will be developed beyond the current state-of-the-art. ACSEPT will also pave the way towards more integration between partitioning and transmutation by carrying out dissolution as well as actinide conversion studies. All experimental results will be integrated by carrying out engineering and systems studies on aqueous and dry (pyro) processes to prepare for future demonstration at a pilot level.

GETMAT is a five-year project on materials that started on 1 March 2008. This cross-cutting project characterises materials for nuclear applications in the area of fission and fusion. Based on the analysis of running projects on innovative reactor and transmutation systems and the needs in terms of structural materials assessment, the focus of GETMAT has been put on ferritic-martensitic (F/M) and oxide-dispersion strengthened (ODS) steels, as cross-cutting structural material choice for core and primary components. Moreover, the GETMAT project aims to streamline and integrate the R&D effort of the European materials laboratories for the performance of the above classes of alloys, in terms of: *i)* availability, fabricability and fundamental properties; *ii)* compatibility with coolants; *iii)* response to irradiation; *iv)* an effort to understand the physical reasons for their behaviour under these conditions. Priorities and objectives GETMAT are: *i)* improvement and extension of 9-12 Cr F/M steels qualification; *ii)* ODS alloy development and characterisation; *iii)* joining and welding procedures qualification (relevant for both ODS and F/M steels); *iv)* development and definition of corrosion protection barriers; *v)* improved modelling and experimental validation.

EUFRAT is a four-year transnational access project at the European Commission, Joint Research Centre (JRC), Geel, Belgium that started on 1 November 2008. Access of outside users to the GELINA and the Van de Graaff accelerator facilities of the Neutron Physics unit of JRC is facilitated for neutron cross-section measurements. The project also promotes a coherent use of the measurement infrastructure in order to meet high-priority neutron data requests from European industry, safety authorities and the nuclear research community. Proposals for experiments submitted by external users are evaluated by a Programme Advisory Committee composed of high-level experts belonging to relevant international organisations. The project is expected to deliver new, more accurate neutron cross-section data in nuclear technology domains such as fission reactor technology, fission reactor and fuel cycle safety, high burn-up fuels, nuclear waste transmutation and innovative reactor systems.

The Integrated Infrastructure Initiative (I3) ACTINET-I3 project is under negotiation. Its aim is to reinforce the networking of existing European infrastructures in actinide sciences, and to facilitate their efficient use by the European scientific community. The objectives of ACTINET-I3 are: *i)* to establish a network of actinide facilities across the EU to integrate and structure the way these actinide infrastructures operate and to foster their joint development in terms of capacity and performance; *ii)* to support and manage jointly a programme of access to appropriate infrastructures for training and associated research projects making use of the proposed facilities; *iii)* to conduct on a limited scale a set of Joint Research Activities (JRA) involving member organisations, with an objective to improve the performance of infrastructures by developing new relevant instrumentations and/or data of common interest. Further, these activities will be complemented by a virtual infrastructure providing a limited support in theory and modelling, with a focus on the complementarities between theory and experiments.

The project on Fabrication, Irradiation and Reprocessing of Fuels (FAIRFUELS) and targets for transmutation is under negotiations. It aims to provide a way towards a more efficient use of fissile material in nuclear reactors. Its objective is to reduce the volume and hazard of high-level long-lived radioactive waste. In fabrication technology and assessment of transmutation performance, FAIRFUELS will focus on minor actinides. Dedicated fuels will be fabricated and a comprehensive irradiation programme will be carried out to address transmutation performance. Certain post-irradiation examinations (PIE) of existing fuels and targets will also be conducted to provide in-depth information on their irradiation behaviour. In support of the PIE, modelling aspects of these fuels will be developed. A training and education programme will also be implemented.

The establishment of a Central Design Team (CDT) for the design of a fast spectrum transmutation experimental facility (FASTEF) working in subcritical mode (ADS) and/or critical mode able to demonstrate efficient transmutation and associated technology is considered as an essential next step just when the FP6-EUROTRANS (see above) is completing its work successfully. There is also a clear need of an alternative technology to sodium. Therefore, FASTEF is proposed to be designed to an advanced level using liquid lead technology. A possible decision to embark upon its construction is expected at the horizon of 2012 with the following objectives: to demonstrate the ADS technology and the efficient transmutation of high-level waste; to operate as a flexible irradiation facility; to contribute to the demonstration of lead fast reactor technology without jeopardising the above objectives. The work to be carried out involves a definition of specifications of FASTEF and an advanced design of the facility in subcritical and critical mode including plant requirements and a study of key issues towards the realisation of such a facility including site specifications and licensing issues.

7 Conclusions

The second tranche of the funding decision of projects for research and training activities in the field of partitioning and transmutation under the Euratom 7th Framework Programme has been completed (see Table 3) whereas a call for proposals for the third tranche has been scheduled to be made in November 2008. Three P&T projects in FP7 have already started and three are under negotiation. Several projects of the FP6 have been successfully completed and the remaining ones are due to be completed in a year or two (see Table 1).

Significant progress has already been made in FP6 in establishing the European Research Area in P&T in the EU and it will be further reinforced in FP7. A well-balanced portfolio of 11 P&T projects in Euratom FP6 (2002-06) were funded with a total budget of ~81 M€ and a EC contribution of ~43.5 M€. All major actors of P&T in EU are a part of these projects. Six P&T projects under FP7 (2007-11) have already been funded with a total budget of ~56 M€ and EC contribution of ~25 M€. Several additional calls for proposals are planned under FP7.

The EU strategy in the area of P&T is well enumerated in Section 2. Implementation of this strategy by projects in FP6 and FP7 is detailed in Sections 4 and 6. It is further recommended that dedicated efforts should be made in developing and selecting low activation materials in reducing the intermediate-level waste and reducing secondary waste streams in the processes of P&T so that it does not put undue burden on the safe disposal of additional secondary waste produced. Moreover, keeping in mind the natural decay of nuclear waste, a careful roadmap and planning (taking account of the time needed *e.g.* for regulatory authority approvals) should be made so that there is no mismatch between the schedules of partitioning, transmutation and disposal technologies. The geological disposal community should further take into account the requirements and accommodate the waste streams emanating from the advanced minor actinide reprocessing systems and support development of appropriate waste forms for geological disposal.

Acknowledgements

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Special session

Fuel cycle strategies and transition scenarios

Chairs: M. Salvatores, K. Ono

US activities on fuel cycle transition scenarios

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Abstract

Countries with active nuclear programmes typically have as a goal transitioning to a closed fuel cycle. A closed fuel cycle enables long-term sustainability, provides waste management benefits, and as a system, can reduce overall proliferation risk. This transition will take many decades, thus the study of the actual transition is an important topic. The United States systems analysis activities as part of the Advanced Fuel Cycle Initiative (AFCI) provide the integrating analyses for the fuel cycle programme, and recent activities are focusing on transition options, and specifically, the dynamics of the transition.

The United States is still studying both one-tier (recycling in fast reactors only) and two-tier (recycling in both thermal and fast reactors) systems, and the systems analysis activities provide insight into the trade-offs associated with the systems, and variations of each. Most recently, a series of sensitivity studies have been completed which provide insight into the behaviour of a transitioning system. These studies evaluate the impact of changing various parameters in the fuel cycle system, and provide insight into how the system will change as parameters change. Because these deployment analyses look at the development of nuclear energy systems over a long period of time, it is very unlikely that we will accurately predict the system's characteristics over time (for example, growth in electricity demand, how quickly nuclear reactors will be deployed, how many fast rectors versus thermal reactors, the conversion ratio of the fast reactors, etc.). How the system will develop will depend on a variety of factors, ranging from political to technical, rational to irrational. Because we cannot accurately predict the future, we need to understand how things could change, and what impact those changes have.

Analyses of future fuel cycle systems require a number of assumptions. These include growth rates for nuclear energy, general architecture of fuel cycle facilities, when new technologies and facilities will become available, how those technologies and facilities will perform, and what policies will be followed concerning used fuel inventories, etc. Since these assumptions have a significant impact on the analyses, they are carefully developed based on the best available information, including technology performance, policy, regulations and historic trends. Still, they are only assumptions – sensitivity analyses help to understand the impact if the assumptions prove to be inaccurate.

This paper covers the recent systems analysis activities in the US AFCI programme, with a focus on the recent sensitivity studies. It also discusses future activities which will include targeted trade studies to help inform fuel cycle decisions.

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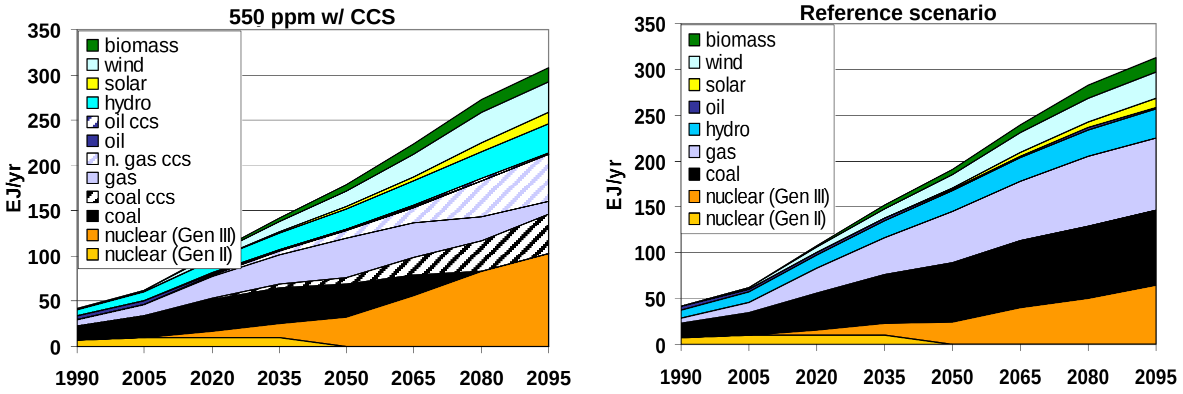
Analyses of future fuel cycle systems require a number of assumptions. These include growth rates for nuclear energy, general architecture of fuel cycle facilities, when new technologies and facilities will become available, how those technologies and facilities will perform, and what policies will be followed concerning used fuel inventories, etc. Since these assumptions have a significant impact on the analyses, they are carefully developed based on the best available information, including technology performance, policy, regulations and historic trends. Still, they are only assumptions – sensitivity analyses help to understand the impact if the assumptions prove to be inaccurate.

The analyses summarised here have highlighted a number of key findings associated with nuclear energy growth and fuel cycle transitioning. To understand the magnitude of potential nuclear energy growth, energy demand and nuclear competitiveness are assessed. Long-range global energy models based on the US Climate Change Science Program Reference Scenario project a five-fold increase in world electricity consumption by the end of the century. The Reference Scenario indicates nuclear energy is competitive with other energy sources and shows an increasing reliance on nuclear energy later in the century, with the world's nuclear share of the electricity market rising from the current 16% to 20% and the United States' share rising from 20% to 25%.

Actions to reduce carbon dioxide emissions to address climate change significantly increase the projected growth of nuclear energy and other low-carbon energy sources. Figure 1 shows the change in sources of electricity generation when a CO_2 emissions constraint is applied globally and carbon capture and storage (CCS) technologies are successfully developed for use with fossil fuels. The Reference Scenario shown in Figure 1 is based on the *Scenario of Greenhouse Gas Emissions and Atmospheric Concentrations* from the US Climate Change Science Program [2], which incorporates the most up-to-date historical information and assumptions regarding future demographics, socioeconomic parameters and energy use, and thus, employs an important reference baseline for long-term global energy and economic analysis. With CCS, nuclear energy market share increases to 33% of electricity generation globally and 41% domestically. Without CCS technologies, nuclear energy market share increases to 50% globally and 58% domestically. These analyses do not consider non-electric applications of nuclear energy that may generate additional growth, such as transportation, water desalination, chemical industry, direct steam used for district heating or resource extraction (tar sands).

While these analyses using the once-through fuel cycle and a global energy simulation indicate the significant value of nuclear energy as one of the key tools for reducing CO_2 emissions to address climate change, they also show huge increases in uranium demand and used fuel inventories. The impact of deploying an advanced fuel cycle to address these system demands is assessed based on transitioning to recycling in the US (global transition studies are planned as a future Systems Analysis activity). The domestic analyses address the system design and deployment challenges for a closed fuel cycle to be economically deployed in the US to meet energy demands while minimising the impact on natural resources and the environment. Analyses consider key issues such as timing, constraints and costs.

Figure 1: Global electricity generation without carbon constraints and with a CO_2 emissions constraint designed to stabilise atmospheric CO_2 at 550 ppm



US energy growth is projected by extending the 25-year growth rates generated by the Energy Information Agency's Annual Energy Outlook reports, while nuclear growth is nominally assumed to be 1.75%/year (starting in 2015), which is sufficient to achieve 28% electricity market share by the end of the century (this US market share is similar to that projected by the global model Reference Scenario, which calculates growth rates dynamically based on demand curves and competing energy technologies. Sensitivity studies also assess higher and lower growth rates and associated market shares). With projected nuclear energy growth in the US, a once-through fuel cycle will require ~4 200 000 tonnes of additional uranium by the end of the century and generate an additional ~410 000 tonnes of used fuel, including 5 300 tonnes of transuranics (TRU) needing geologic disposal.

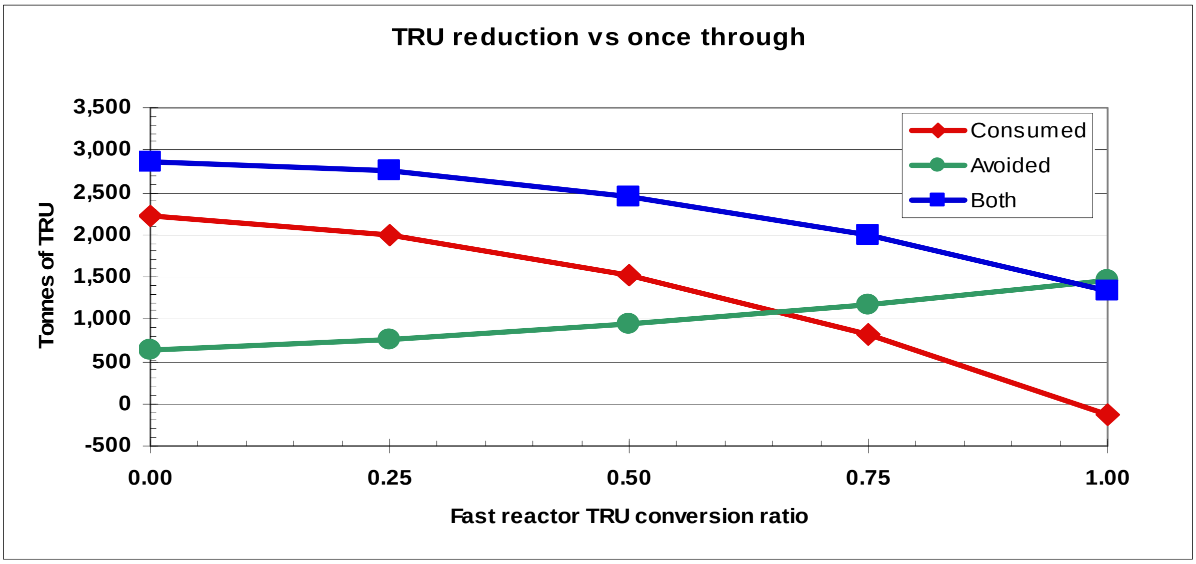
The impact of advanced fuel cycle technologies on these numbers is highly dependent on the rate of deployment of separations facilities and fast reactors. The size and timing of uranium oxide (UOX) used fuel separations facilities is the primary driver behind the rate of deployment of TRU-burning fast reactors. The fast reactor deployment rate in turn impacts the amount of TRU both generated and consumed.

Fast reactor deployment rates will be much lower than the levels predicted by simple "equilibrium" calculations due to multiple system constraints that impact the amount of TRU available for fuelling new reactors at start-up. Sensitivity studies have identified the most important constraints (in order) to be separations capacity and timing, nuclear growth rate, fast reactor TRU conversion ratio (the ratio of the mass of transuranic isotopes created over those destroyed during fuel irradiation; fast reactors can operate with TRU conversion ratios both below and above 1.0.), used fuel cooling time, and fast reactor introduction timing. In a one-tier system (where recycling takes place in fast reactors only) with fast reactors at a TRU conversion ratio of 0.5, over 75% of the reactor fleet will still be LWR in 2100. A two-tier system (where recycling takes place first in thermal reactors, then in fast reactors) with a mixed-oxide uranium/plutonium fuel (MOX-Pu) recycle stage in LWR imposes additional constraints that result in over 90% LWR in 2100. These additional constraints are related to the time required for the MOX-Pu cycle, including both fuel irradiation and cooling.

Transuranics are important for two reasons: they can substitute for uranium in fuel, and they are the primary contributors to long-term waste management impacts. TRU management must account for both the TRU consumed in fast reactors and the additional TRU generation avoided due to fast reactors replacing some LWR. Each factor reduces TRU quantities by over 1 000 tonnes during the century (versus once-through). The more TRU "at work" in fast reactors, the more total TRU is reduced. Figure 2 shows the impact on TRU management as a function of fast reactor transuranic conversion ratio.

The early phases of transitioning from once-through to a closed fuel cycle require extra flexibility to address uncertainty in initial facility sizes and timing that can drive material supply/demand mismatches. For example, a later date for initial fast reactor deployment in a one-tier system can result in large inventories of separated TRU requiring secure storage, unless MOX-Pu is used in LWR (an interim two-tier system) or separations deployment is also later (which moves the system back toward the status quo of excess used fuel in storage).

Figure 2: Transuranics reduction between 2000 and 2100 with a one-tier fuel cycle as a function of conversion ratio



The location of used fast reactor fuel recycling facilities (on-site versus centralised) also has a significant impact on the rate of fast reactor deployment, impacting the amount of TRU tied up in used fuel due to the added cooling time required for efficient transportation (transportation of used fuel is often decay-heat-limited, with minimum cooling times required to ship full casks).

The costs associated with facility construction and operations for the once-through, one-tier and two-tier fuel cycles were analysed to determine both total costs and the relative share for each system coming from the fuel cycle front end and back end and the reactors. A closed fuel cycle employing advanced technologies is projected to increase the total system cost of nuclear-generated electricity by only 5 to 6 $/MWh (~10%), but the cost uncertainties are large. The primary contributor to the increased cost is the projected capital cost of fast reactors, with fuel recycling costs contributing between 20 to 40% of the total cost difference.

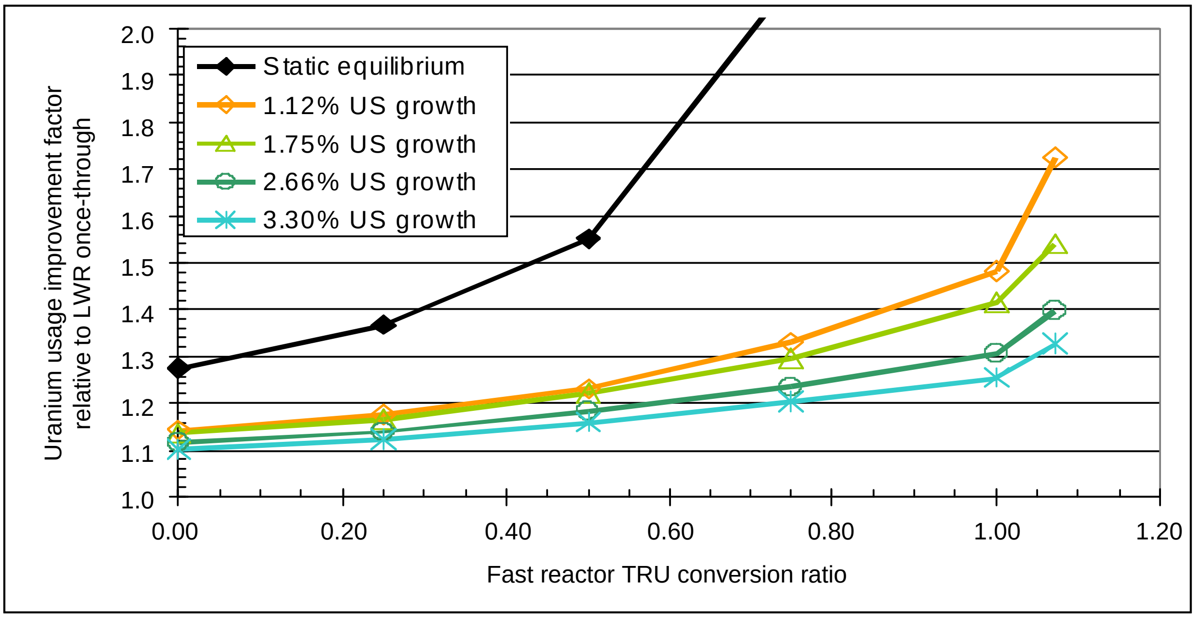
Costs were also compared to competing base-load electricity generation systems. With either an open or closed fuel cycle, nuclear costs are projected to be competitive with fossil fuels even without a carbon tax, and the total cost uncertainties for nuclear (although large) are less than the cost uncertainties of fossil fuel energy systems.

The once-through fuel cycle consumes less than 1% of mined uranium, with the rest left in enrichment tails and used fuel. Recycling of used fuel allows for slightly more of the uranium to be consumed, but 100% of the depleted uranium from enrichment and 99% of the uranium recovered from used fuel remains unused at a fast reactor conversion ratio of 0.50.

Dynamic analyses show that transitioning to a closed fuel cycle only reduces uranium needs in this century by ~750 000 tonnes (~18%), assuming fast reactors with a TRU conversion ratio of 0.5.

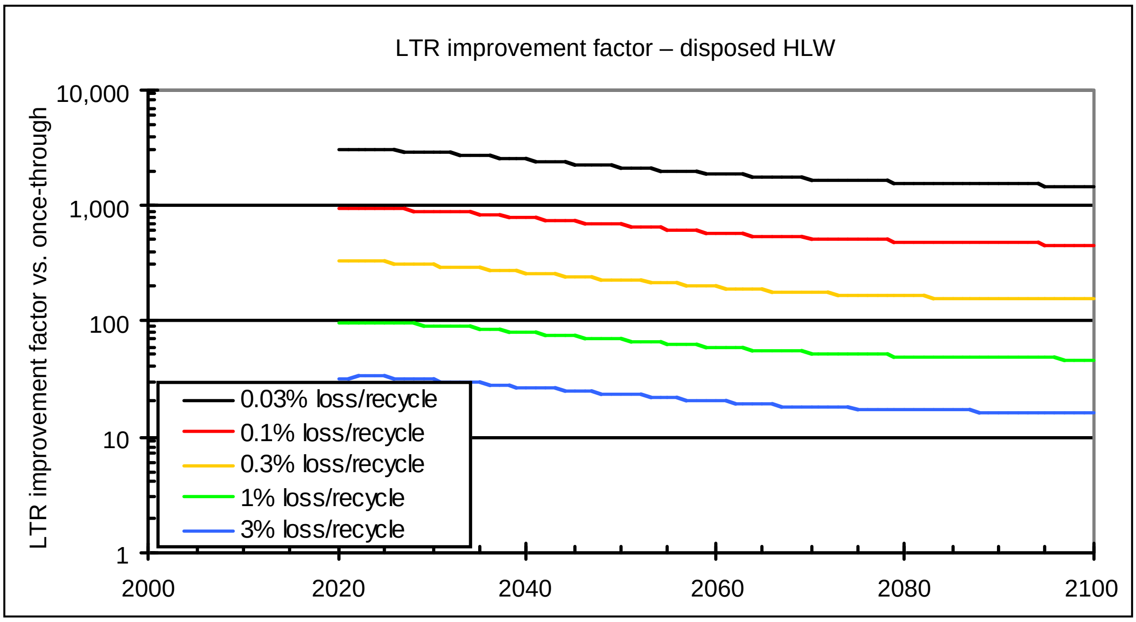
Figure 3 shows the combined impact of growth rate and conversion ratio on uranium consumption by the end of the century. Uranium usage is improved when higher conversion ratio fast reactors are deployed, but large savings will not occur until the next century unless fast reactor conversion ratios are substantially greater than 1. This figure also shows the large difference in results obtained when dynamic analysis is used versus static analysis. Dynamic analysis includes facility life, start-up material requirements and other factors that constrain the rate of transition of systems. These constraints are not considered in static analysis, often resulting in larger overestimates of the near-term impact of closed fuel cycle systems.

Figure 3: Static and dynamic analysis of uranium usage improvement factors relative to once-through as a function of both growth rate and conversion ratio



The impact of transitioning to a closed fuel cycle on waste management is large. Sensitivity analyses on the efficiency of recycling facilities show both the long-term radiotoxicity and long-term decay heat of waste from recycled fuel needing geologic disposal reduced by more than a factor of 100 versus once-through if uranium and TRU losses to waste could be held below 0.5% (see Figure 4). These improvements reduce the time the waste is more radiotoxic than uranium ore from ~300 000 years to ~300 years, while making space-efficient geologic disposal much easier.

Figure 4: LTR improvement factors for one-tier versus once-through at different recycle efficiences



The analyses summarised here provide answers to a number of questions concerning the transition to an advanced fuel cycle, and indicate areas where future studies would be useful:

- Assessing the impact of advanced fuel cycle cost differentials on domestic and global projections of nuclear energy growth.
- Assessing phased fuel cycle transition options, including the initial fielding of mature technologies followed by a later phase-in of advanced technologies.
- Supporting major technology decisions and requirements development through integrated analyses such as:
  - minor actinides storage vs. disposal trade-off study;
  - system losses trade-off study;
  - waste trade-off studies.
- Extending the types and scope of analyses provided.
- Impacts of expansion of nuclear energy beyond electricity generation.

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Current status of the Japanese nuclear fuel cycle – outline of the Japanese nuclear fuel cycle strategy and transition scenario to FR cycle

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Abstract

The G8 leaders declared, at the Hokkaido Toyako Summit in 2008, "to seek to share with all Parties to the UNFCC the vision of, and together with them to consider and adopt in the UNFCCC negotiations, the goal of achieving at least 50% reduction of global emissions by 2050" [1]. They also referred to nuclear energy as "a means to addressing climate change and energy security concerns and as an essential instrument in reducing dependence on fossil fuels and hence green gas emissions in a growing number of countries."

Japan has been aggressively utilising nuclear energy as one of the major electric power sources to fulfil its domestic energy demand, by currently operating more than 50 nuclear power plants with the total capacity of ca. 50 GWe. This energy source highly depends on the imports of natural uranium from foreign countries to supply fuel to the nuclear power plants. Since nuclear energy use is growing throughout the world; meaning much more uranium resources are to be required in the future, it is rather difficult for Japan alone to maintain the stable supply of uranium resources on a longer-term range. Therefore, the establishment of nuclear fuel cycle with plutonium recycling is indispensable to realise the efficient use of natural uranium resources. Moreover, commercialisation of fast reactors and related fuel cycle (FR cycle) system is aimed at improving drastically the efficiency of natural uranium utilisation and at the same time minimising our environmental concern on geological disposal of high-level radioactive waste. The establishment of closed FR cycle technologies is hoped to secure the long-term sustainable and stable energy supply and to help prevent global warming through reduction of greenhouse gas emissions.

This paper describes the current status of nuclear energy and the scenario for the sustainable nuclear energy in Japan.

Current status of nuclear power plants in Japan

Considering the expected increase in Japanese electricity demand in the future, the electricity supplied by nuclear power plants is planned to expand further. Table 1 shows the status of nuclear power plants currently operating or planned in Japan [2]. A fleet of 55 commercial nuclear power plants, consisting of 23 pressurised water reactors and 32 boiling water reactors, are in operation with a total generation capacity of ca. 50 GWe, supplying over 30% of the domestic electricity demand. The nuclear energy will reach 68 units including new builds under construction or planned with a total capacity of 66.7 GWe in 2017, which corresponds to 41.5% of domestic electricity demand.

Table 1: Current status of nuclear power plant in Japan

Number of NPP (March 2007)	55 (23 PWR and 32 BWR)
Installed capacity (March 2007)	49.6 GW
Electricity generation (2006)	304.5 TWh
New builds (under construction or planned)	13
Total capacity with new builds (by 2017)	66.7 GW

Concerning the nuclear fuel cycle facilities in Rokkasho, the commercial reprocessing plant is about to be commissioned in 2009, in addition to the Rokkasho enrichment plant which has already been in operation and a MOX fuel fabrication plant being planned. Moreover, an Ohma ABWR with a full MOX core is under construction with a scheduled start-up in 2012.

From the viewpoint of preservation of the global environment, nuclear energy contributes to save about 68 million tonnes of oil per year and about 240 million tonnes (carbon equivalent) of CO_2 emission per year. Thus an expanded use of the nuclear energy is essential to meet electricity demand while effectively cutting CO_2 emission by 50% in 2050 in accordance with the declaration of the Hokkaido Toyako Summit.

Development plan of FR cycle system in Japan

A “Feasibility Study (FS) on Commercialised FR Cycle Systems” was completed in 2006. In the FS, the combination of a sodium-cooled fast reactor with oxide fuel, advanced aqueous reprocessing and simplified pelletising fuel fabrication was selected as a main concept which should be developed principally. Subsequently, a new Fast Reactor Cycle Technology Development (FaCT) project was launched in Japan focusing on development of the main concept. Figure 1 shows an outline of development plan toward commercialisation of FR cycle technology in Japan [3,4]. A conceptual design study of the main concept and the R&D programmes of innovative technologies to be adopted in the main concept are being conducted toward an important milestone at 2015. Thereafter, the FR cycle development project will enter into an introduction stage through system demonstration. The demonstration FR cycle will start to operate around 2025. The commercialised FR cycle system will be deployed starting at around 2050 based on the experience of the demonstration FR cycle system.

In the FaCT project, the R&D activities are being carried out with the development targets (or objectives) and the design requirements in which the development targets are embodied. These development targets defined in the FaCT project are listed in Table 2. The four development targets are: *i)* safety and reliability; *ii)* sustainability’; *iii)* economic competitiveness; *iv)* nuclear non-proliferation. The targets are defined, and the sustainability target is further sub-categorised into environmental protection, waste management and efficient utilisation of nuclear fuel resources. These development targets are further elaborated in the design requirements for both the reactor and fuel cycle systems.

For the reactor system in the FaCT project, the Japan Sodium-cooled Fast Reactor (JSFR), a large commercial-scale reactor, has been conceptually designed with a generation capacity of 1.5 GWe, an advanced loop-type cooling system and a MOX-fuelled core. A total of 13 new innovative technologies were identified and included in the current design in order to satisfy the stringent design requirements. They are categorised into three areas: economic competitiveness, enhancement of reliability and enhancement of safety. The innovative technologies with regard to economic competitiveness include: shortening of piping length by adoption of high chromium steel, reduction of the loop number to

Figure 1: Outline of development plan toward commercialisation of FR cycle technology in Japan

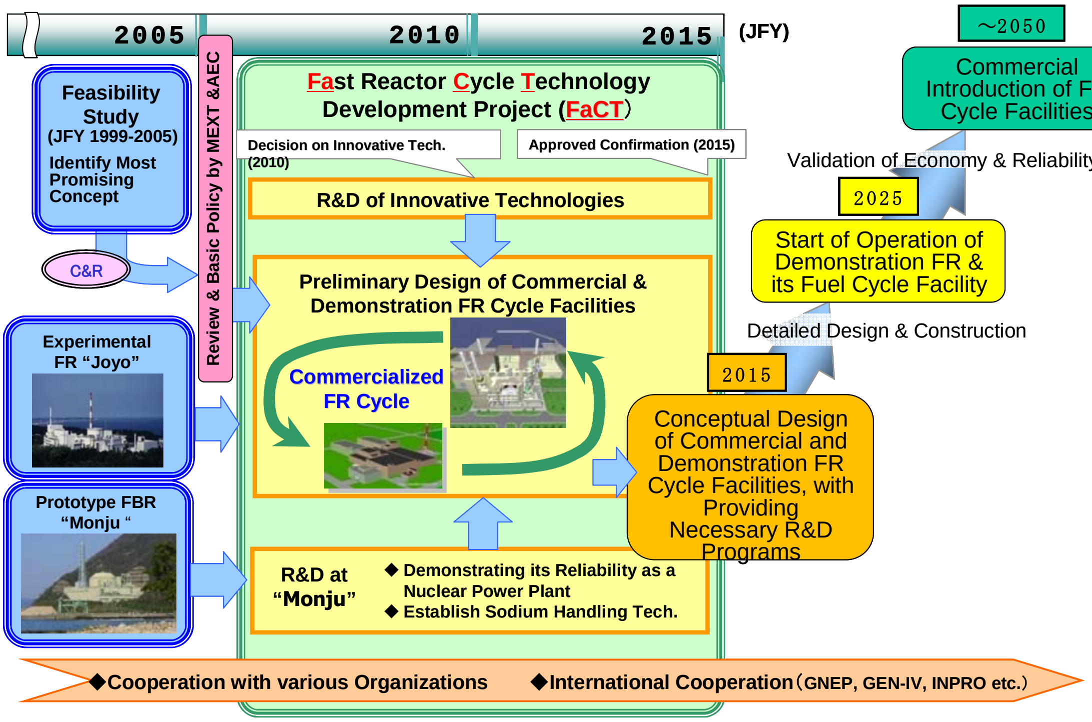


Table 2: Development targets of FaCT project

Safety and reliability	
SR-1	Ensuring safety equal to future LWR and related fuel cycle facilities
SR-2	Ensuring reliability equal to future LWR and related fuel cycle facilities
Sustainability	
Environmental protection	
EP-1	Radioactive influence through normal operation no more than future LWR cycle
EP-2	Emission control of environment transfer substances which can restrict in safety limits
Waste management	
WM-1	Reduction of an amount of radioactive waste compared with future LWR cycle
WM-2	Improvement of waste management ability equal or more than future LWR cycle
WM-3	Reduction of radiotoxicity compared with future LWR cycle
Efficient utilisation of nuclear fuel resources	
UR-1	Breeding ratio to enable transition to fast reactor, and its flexibility
Economic competitiveness	
EC-1	Electric generation cost which can match other power plants
EC-2	Investment risks no more than future LWR cycle
EC-3	External costs no more than future LWR cycle
Nuclear non-proliferation	
NP-1	Adoption of institutional measures and application of technical features which can enhance non-proliferation
NP-2	System design of physical protection and its development

two-loop system, integration of IHX with primary pump, compact reactor vessel using a hot-vessel concept, high burn-up core using ODS (oxide-dispersion strengthened steel) cladding, simplified fuel handling system, and simplified containment vessel with a steel-concrete composite structure. The innovative technologies on enhancement of reliability includes: highly reliable SG with double-wall tubes, a double-boundary system for sodium piping, and in-service inspection and repair technology such as under-sodium viewer.

For the fuel cycle system, the combination of the advanced aqueous reprocessing system and the simplified palletising fuel fabrication system has been identified as the main concept. The advanced aqueous reprocessing system has a significant advantage of almost halving the construction cost compared with the conventional technology; this is to be realised by streamlining the installations and lowering a decontamination factor by eliminating some processes such as a complicated purification system. Meeting the development targets of sustainability and nuclear non-proliferation is evident as well mainly by adopting the low decontamination fuel with full TRU recycling.

After launching the FaCT project, a government-level, five-party council, consisting of MEXT, METI, electricity utilities, manufacturers and JAEA, was established to develop a roadmap for the FR cycle development programme towards commercialisation of FR cycle technology in Japan. The roadmap will make it possible to promote effective and efficient R&D activities over a long time and to facilitate smooth transition through the demonstration stage. It will also specify the requirements for both the demonstration and commercial facilities, for the former of which the conceptual designs are to be proposed in 2015. This is then followed by the activities in demonstration and commercialisation stages until 2025 including a plan for constructing demonstration facilities.

For the reactor system development, Mitsubishi Heavy Industries (MHI) was selected as a “core” enterprise of FR development programme for commercialisation in April 2007, and MHI then has established a new engineering company, Mitsubishi FBR Systems, Inc. [5]. Regarding the fuel cycle technology, since a next, post-Rokkasho reprocessing plant will play an essential role to realise a longer-term fuel cycle technology in Japan, a new governmental working group was formed under the above-mentioned five-party council to discuss possible next-generation reprocessing technologies. The FR deployment scenario study described in the next section was presented at the working group to provide a basis of the requirements for future technology to be developed in Japan.

FR deployment scenario studies

In this section, the Japanese typical FR deployment scenarios are described [6,7]. It is required to keep sufficient flexibility to accommodate future uncertainties such as a level of nuclear power plant capacity, timing of FR deployment, etc. and to achieve smooth transition from LWR to FR fuel cycle. The objective of the FR deployment scenario studies therefore is to seize the characteristics of FR and related fuel cycle, and the specifications of the second (post-Rokkasho) reprocessing (for both LWR-SF and FR-SF) plant.

Assumptions

The basic boundary conditions for the present FR deployment scenario study are as follows:

- transition from LWR cycle to FR cycle with keeping Pu balance;
- upper limit of Pu storage in Rokkasho plant: 20 tonnes Puf;
- average load factor of the second reprocessing plant: over 80%.

Two cases are considered for the nuclear power generation capacities as shown in Figure 2. The reference case is a low-growth scenario where the nuclear power generation capacity is assumed to increase to 58 GWe by 2030 and to be kept constant thereafter. In the expansion case, it is assumed the nuclear power capacity increases to about 110 GWe by 2060 and is kept constant thereafter. The latter case assumed a great shift from fossil fuel to electricity in the field of heat utilisation in the civilian sector as a global warming countermeasure and continuance of transition from fossil fuel to electricity in the industrial sector (60% electrification ratio at 2100).

Other assumptions for the characteristics of the reactor system (LWR and FR) and the nuclear fuel cycle system are shown in Table 3. The breeding ratios of FR are 1.03 for low breeding core and 1.2 for high breeding core. The fuel burn-ups of LWR and FR are assumed to be 40-60 GWd/t and 150 GWd/t (core fuel), respectively. Plant lifetimes are assumed to be 60 years for reactors and 40 years for reprocessing plants. The minimum ex-core time periods are four years for LWR cycle and five years for FR cycle. Even though a 10% loss rate for Np is taken into consideration, the whole actinide loss rate of FR cycle is smaller than that of LWR cycle (1%).

Figure 2: Nuclear power generation capacity of the reference case and the expansion case

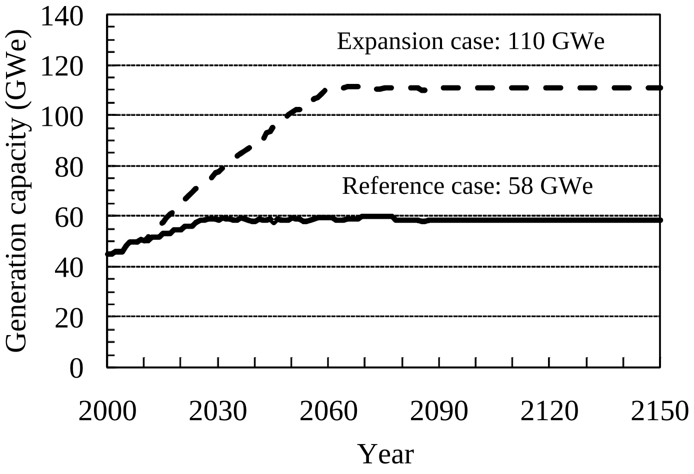


Table 3: Outline of characteristics for facilities

Items		Outline of characteristics
Nuclear power plant capacity (previous slide)		58 GWe (reference case) 110 GWe (nuclear energy use expansion case)
Reactor systems	LWR	-2029: burn-up 40 000 MWd/t, load factor 80% 2030-: burn-up 60 000 MWd/t, load factor 90% LWR-MOX: 2010-2045 (about 30 years, keeping with Pu balance)
	FR	Breeding core (BR = 1.2), break-even core type (BR = 1.03) Replace all LWR to FBR after FR deployment in 2050 (or 2040)
Fuel cycle systems	Ex-core time period	LWR: 4 years, FR: 5 years
	Loss factor	LWR (uranium conversion: 0.5%, fabrication: 0.1%, reprocessing: U – 0.4%, Pu – 0.5%, MA – 0.1% FR (fabrication: 0.1%, reprocessing: U/Pu/Am/Cm – 0.1%, Np – 10%)
	Other	RRP (Rokkasho reprocessing plant): plan in Sept. 2007 FR reprocessing: demonstration plant, 20 tHM/year, 2030 in operation LWR cycle: recovered uranium usage, MA recovery starts from 2nd reprocessing plant MA burning in FR (upper limit 5%) Plant lifetime: 60 years for reactors, 40 years for fuel cycle facilities

Effects of nuclear power plant capacities

In this scenario study, the nuclear power generation capacities and the reprocessing capacities were evaluated quantitatively for both the reference and expansion cases. Figures 3 and 4 show the nuclear power generation capacities for each reactor type of the reference and expansion cases, respectively. Commonly to the two cases all LWR will be replaced by FR by within 60 years starting from 2050 until 2110. Deployment of the low-breeding type FR, which is superior in economic competitiveness, will start from 2065 in the reference case and from 2090 in the expansion case. This is because twice as many FR need to be deployed in the expansion case, requiring high breeding type FR initially and for a longer time period.

Figure 5 shows the LWR reprocessing capacity. In the expansion case, the second LWR reprocessing of a 1 200 tonnes/year capacity is started up in 2042. The total required capacity of reprocessing in the Rokkasho and the second LWR reprocessing plant will be 2 000 tonnes/year for the five years until 2046. Further, the capacity of 2 400 tonnes/year will be needed for 20 years (from 2062 to 2081) by the second LWR reprocessing plant and the third LWR reprocessing plant to be operated simultaneously.

Figure 3: Nuclear power plant capacity for reference case

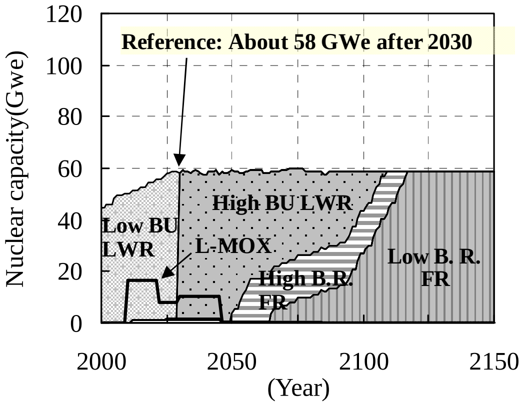


Figure 4: Nuclear power plant capacity for expansion case

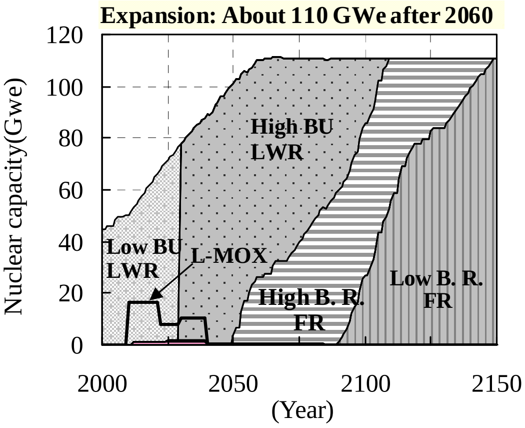
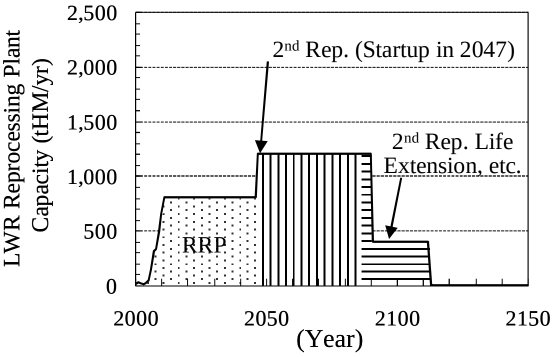


Figure 5: LWR reprocessing capacity

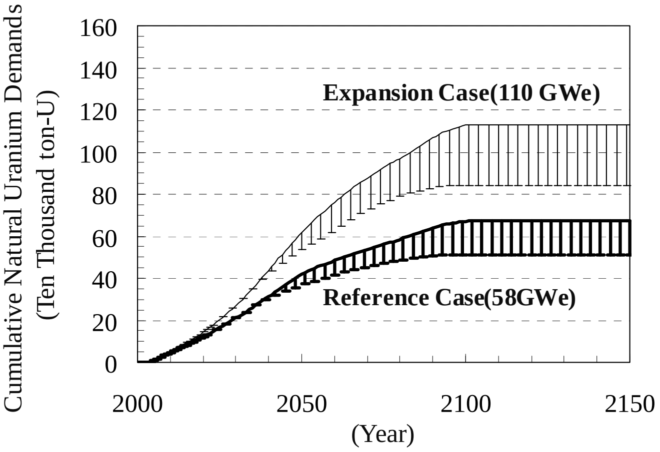


Effects of FR Deployment

A FR deployment scenario should be discussed from various aspects such as: efficient utilisation of nuclear fuel resources, waste management, economic competitiveness, etc. Efficient utilisation of nuclear fuel resources and waste management are considered as one of the largest advantages to realise JSFR and related fuel cycle. In this section, a typical FR deployment scenario is reviewed from these viewpoints [8].

First, the cumulative natural uranium demands with FR deployment in the reference and expansion cases are shown in Figure 6, where the upper and lower bounds in each case correspond to the FR deployment in 2050 and 2040, respectively. The uranium demands will reach saturation after the completion of switchover to FR; in other words, the cumulative uranium demand of Japan continues to increase monotonically without FR deployment. The cumulative uranium demands until the completion of switchover to FRs were 670 thousand tonnes for the reference case and 1 130 thousand tons for the expansion case for the 2050 FR deployment scenario. The effect of early FR deployment (2040) was evident, significantly decreasing the uranium demands to 510 and 840 thousand tonnes for the reference and expansion cases, respectively.

Figure 6: Cumulative natural uranium demand of Japan



The second aspect of our scenario study concerns the amount of radioactive wastes generated throughout the fuel cycle. The radioactive waste generation from Japanese nuclear fuel cycle system with FR deployment is shown in Figure 7. The three major peaks appearing in the figure correspond to the time periods when large numbers of plant decommissioning for reactors and reprocessing plants. It is also understood from the figure that the total volume of radioactive wastes is dominated by the low-level wastes (LLW) from nuclear energy system. The advantage of FR deployment is also evident in that radioactive waste generation is reduced significantly as a lesser volume of wastes are generated from decommissioning of the compact and hence economical FR and related fuel cycle facilities assumed in this study. The quantity of high-level radioactive wastes (HLW) per year decreases after the switchover to FR. At the same time, almost all the spent fuels can be reprocessed without interim storage (after 2047). The main reasons for this reduction in HLWs with FR deployment are: the thermal efficiency improvement of FR and the recovery and recycling of minor actinides in the future reprocessing plants. Another advantage of deployment of FR and related fuel cycle is the significant reduction of potential radioactive hazard associated with HLW. Figure 8 shows that the major reduction of potential radioactive hazard when the FR is deployed. The main reason for the decrease of the hazard around 1 000 years after HLW disposal is the minor actinide recovery from raffinate in the reprocessing plants for FR spent fuels.

The third and probably most important aspect is the economic competitiveness which is one of the development targets. In this study, we have evaluated the total electricity generation cost, including essentially all the costs of plant construction, fuel fabrication, spent fuel reprocessing, waste management and disposal, material transport and storage, etc. The conditions used in the analyses were set based on the report of the Advisory Committee for Natural Resources and Energy's subcommittee to study costs and other issues [9]. The evaluated cost is shown in Figure 9 for the cases without reprocessing (LWR once-through) and with FR deployment (reference case). Cash outflow peaks appear in the left figure during 2000s, 2040s-2060s, 2080s-2120s and 2160s. These cost peaks are caused by the construction costs and decommissioning costs of both LWR spent fuel reprocessing plants and the nuclear power plants. The average power generation cost from 2008 to 2200 was calculated from the cash outflows of the Japanese nuclear fleet; which was about 3.9 JPY/kWh with the cost suppositions in this analysis.

Figure 7: HLW generation from nuclear energy with FR deployment in Japan

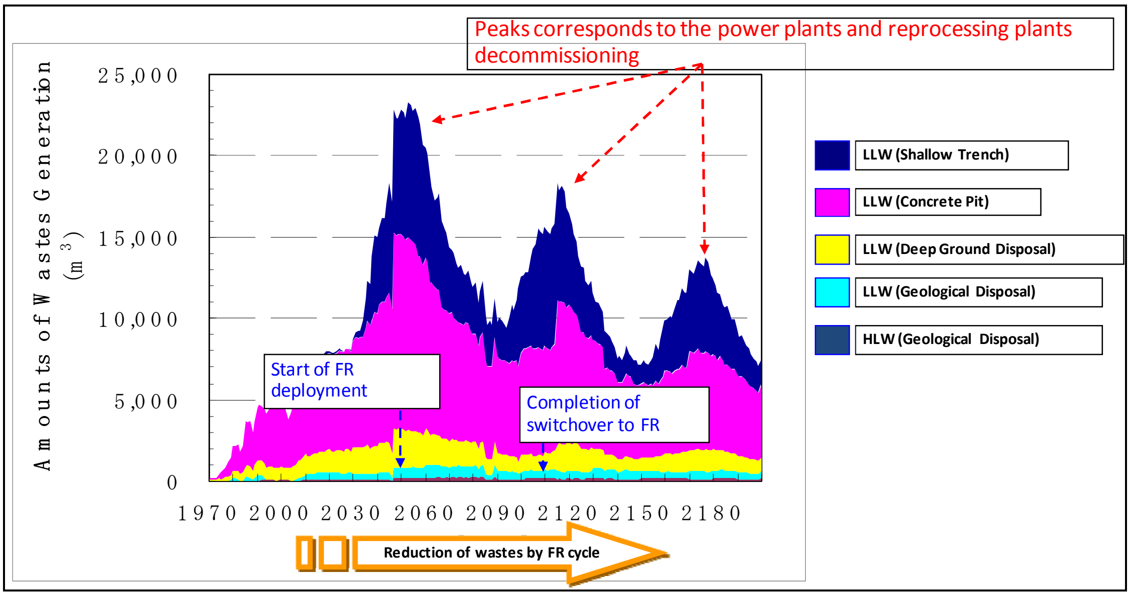


Figure 8: Reduction of potential radioactive hazard with FR deployment

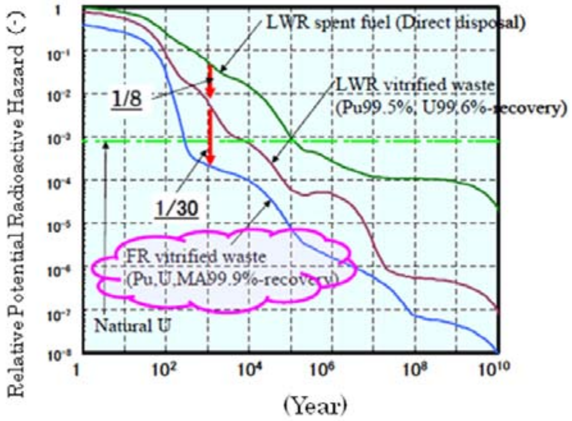
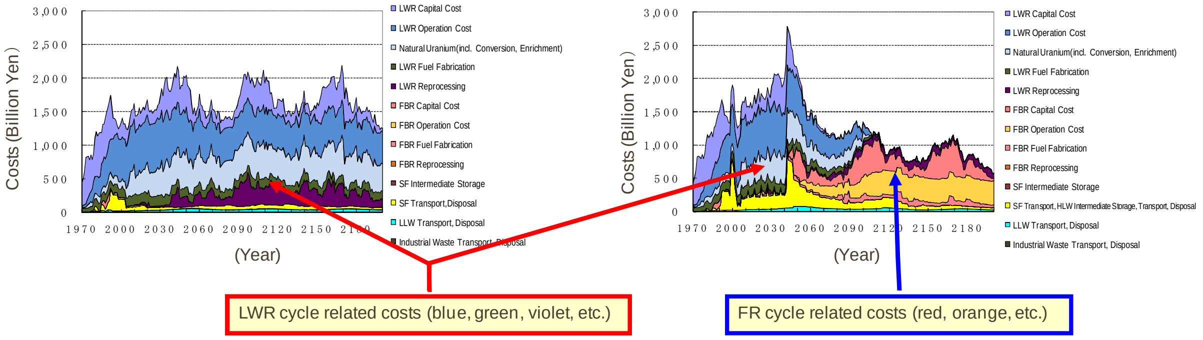


Figure 9: Long-term economics of nuclear energy in Japan

Left: once-through, right: FR deployment



If the economics evaluation result of the FR deployment scenario is compared to that of the once-through scenario (see also Figure 8), the fuel cycle cost related to LWR made the total electricity generation cost of FR deployment scenario higher. However, after the deployment of the compact and economic facilities with innovative technologies and materials related to FR and its fuel cycle, the total cost will drastically decrease. Therefore, the present results imply that the early transition to the FR and their fuel cycle system from the current LWR cycle system is important as well as the high performance levels of the system.

Conclusions

As a country with highly developed industry and poor natural resources, Japan places great emphasis on economic prosperity through securing a stable and sustainable energy supply. In addition, the prevention of global warming has become a common world-wide interest. Japan therefore pursues the use of nuclear energy as a major means to combine economic prosperity and global environmental conservation. Since the FR and related fuel cycle system is capable of extending the available period of nuclear energy drastically, it can be regarded as one of the indispensable pillars of a long-term nuclear energy policy.

The Japanese FR deployment scenarios have been studied to quantitatively evaluate the transitional process toward the commercialisation and deployment of FR. The FR are expected to replace all LWR in Japan within 60 years even if the nuclear power plant capacity is expanded in the future. Although the second reprocessing plant of a 1 200 tHM/y capacity is sufficient to process all the spent fuel, the reprocessing plant should come into operation well ahead of the schedule if the timing of FR deployment occurred sooner than 2050.

The present scenario study also indicated that the deployment of FR is beneficial to:

- reduce the cumulative natural uranium demand for Japan and contribute to sustainable use of nuclear energy in the world;
- utilise HLW disposal site efficiently through reducing the quantities of HLW generation as a result of minor actinide recovery and recycling from the reprocessing plants;
- lower the long-term electricity generation cost of the nuclear energy fleet of Japan if the innovative technologies are installed with the FR facilities.

We believe therefore that the commercialisation of FR and related fuel cycle systems is one of the most important targets for future Japanese society if we take full advantage of the benefits from FR deployment. The JAEA and electricity utilities will jointly develop JSFR and related fuel cycle technologies proactively in the FaCT project to deploy a demonstration FR around 2025 and a commercialised FR around 2050.

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Overview of the French fuel cycle strategy and transition scenario studies*

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Abstract

France has been reprocessing the spent fuel of its nuclear energy generating PWR fleet since 1976. This strategy affords retrieving re-usable nuclear materials and segregating fission products with minor actinides (currently) for a safe packaging as vitrified waste and interim storage while awaiting disposal in a geological repository. Plutonium and reprocessed uranium are (partly) recycled once in PWR (respectively as MOX and Rep-UO_2 *fuel) thus reducing by almost 15% needs for uranium and enrichment separative work. Spent MOX fuel subassemblies are stored as a fissile resource for future fast reactors.*

Scenario studies of nuclear power deployment and utilisation of uranium world wide lead to anticipate an industrial deployment of fast neutron reactors around 2040 in the French generating fleet. This sets an overall time frame for research and pilot-scale demonstrations on next generation fast reactors and advanced recycling modes to prepare their industrial deployment in time.

Furthermore, the French bill of 28 June 2006 on "A Sustainable Management of Nuclear Materials and Radioactive Waste" institutes a strategy for the nuclear fuel back-end in France with plans to open a high-level long-lived radioactive waste repository by 2025. Moreover, it establishes a link between continuing research on partitioning and transmutation to further decrease the long-term burden of current waste packages, and research on Generation IV fast neutron systems with closed fuel cycles, thus acknowledging future fast power reactors as the most likely nuclear systems to perform transmutation at the industrial scale. This bill calls for identifying by 2012 recycling modes that have the most promising industrial prospects and to proceed with their demonstration in a prototype fast reactor in the 2020s.

Subsequently, French nuclear stakeholders (CEA, AREVA and EDF) are currently conducting active research on advanced fast reactors, fuel and fuel cycle technologies to screen promising design features for a new generation fast nuclear system with enhanced safety and economic competitiveness.

Transitional fuel cycle studies from LWR to fast reactors are conducted with a view to assessing:

- *strategies for managing plutonium stockpiles to deploy fast reactors;*
- *potential benefits of homogeneous or heterogeneous recycling of minor actinides to alleviate long-term radioactive waste burden (decay heat and radiotoxic inventory);*
- *limitations of transmutation strategies if prematurely stopped;*
- *needs for new fuel cycle plants for advanced recycling modes;*
- *strategies for enhancing proliferation resistance of closed fuel cycles;*
- *the overall time line and cost associated with advanced recycling modes.*

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* The full text of this paper was unavailable at the time of publication.

Research and development on advanced partitioning processes (in the hot laboratory Atalante) as well as on co-precipitated and/or minor-actinide bearing fuels (including irradiation tests in Phénix) proceed in line with the national research programme that was conducted from 1991 to 2006 on high-level radioactive waste management. They aim at providing sound scientific and technical bases for decisions to be made in 2012 and developing key technologies for the prototype.

This prototype and the associated facilities to produce its driver fuel (MOX, possibly co-precipitated) and experimental minor actinide bearing fuel provide an opportunity to demonstrate all of the steps of a sustainable fuel cycle at a pre-industrial stage in the 2020s. Other initiatives such as the Global Actinide Cycle International Demonstration (GACID) conducted in Japan, the United States and France, are additional essential steps to acquire sound scientific bases and build consensus on international safety and security standards that will apply to future nuclear systems and associated fuel cycle services in national or international centres.

Fuel cycle synergies and regional scenarios

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Abstract

The regional approach proposed in order to implement advanced fuel cycles in Europe, partially reported on the previous IEMPT meeting, has been completed. The impact of various deployment strategies and policies in different countries is shown. Regional facilities' characteristics and potential deployment schedules are discussed. Waste inventories and characteristics, such as decay heat, are also described. Lessons about fast reactor and ADS characteristics have also been learnt.

Purpose of the study and choice of scenarios

Implementation of P&T and advanced fuel cycles within an original regional approach [1,2] has been considered. In fact, it seems essential to study the possibilities to share fuel cycle facilities and to envisage a concerted use of materials, in order to develop at a regional level a sustainable nuclear energy, with optimised use of resources and investments and waste minimisation in an enhanced proliferation resistant environment.

Regional fuel cycle scenarios definition

The scenarios consider different groups of countries:

- Group A is in a stagnant or phase-out scenario for nuclear energy and has to manage its spent fuel.
- Group B is in a continuation scenario for the nuclear energy and has to optimise the use of its resources of plutonium for the future deployment of fast reactors.
- A subset of Group A, after stagnation, envisages a nuclear renaissance.
- Some countries, initially with no nuclear power plants, decide to go nuclear.

Four different scenarios have been considered. All scenarios make use of fast spectrum reactors, both ADS type (Scenarios 1 and 2) or critical (Scenario 3).

Scenarios 1 and 2 consider the deployment of a group of ADS shared by Countries A and B. The ADS use the plutonium of Group A and transmute the minor actinides of the two groups. The plutonium of Group B is either mono-recycled in PWR and then stored for future deployment of fast reactors (Scenario 1) or is continuously recycled in PWR (Scenario 2). In Scenarios 1 and 2, the deployment of critical fast reactors is not simulated. Scenario 3 considers the deployment of fast reactors in Group B. These fast reactors use the plutonium of Groups A and B and recycle all the minor actinides.

Scenario 4 corresponds to a "renaissance" of nuclear energy in selected countries. Starting from Scenario 3, both Group B and some Group A countries will dispose of fast reactors to handle their own TRU inventories. These scenarios are not intended for a stiff classification of the European countries in a given category, nor are they intended to be an exhaustive list of possible scenarios, but rather as a working tool to make a systematic approach to the subject. The numerical examples have been based on the available information provided by a group of selected European countries (Belgium, Czech Republic, France, Germany, Spain, Sweden and Switzerland), that are representative of the overall present European situation. In these scenarios, the regional facilities to be deployed are:

- the standard spent fuel reprocessing facility for Group A and for Group B;
- the fast reactor fuel fabrication facilities and (if ADS are to be deployed);
- the ADS spent fuel reprocessing facility;
- the ADS fuel fabrication facility;
- the ADS.

All studies have been performed with the CEA COSI code [3]. FZK has provided results for Scenarios 1 and 2 (based on the deployment of ADS) and CEA has focused on Scenarios 3 and 4 (based on the use of critical fast reactors). The scenarios were previously described in Ref. [4], where some preliminary results for Scenarios 3 and 4 were also presented.

Scenarios 1 and 2

These scenarios consider the deployment of a transmuting ADS fleet shared by the countries in Groups *A* (Belgium, Czech Republic, Germany, Spain, Sweden and Switzerland) and *B* (France). In these scenarios the ADS will use the plutonium of Group A and will transmute the minor actinides of the two groups; the plutonium of Group B will be mono-recycled (Scenario 1) or multi-recycled (Scenario 2) in PWR and then stored for future deployment of fast reactors (starting from 2040).

The main objectives of these scenarios are:

- to decrease the stock of spent fuel of countries A down to 0 at the end of the century;
- to stabilise the MA inventory of Group B within the end of the century;
- to investigate the required number of ADS to be deployed;
- to determine the number and capacities of the fuel cycle facilities needed;
- to stabilise the Pu inventory of Group B.

The ADS facility used is the lead-cooled European Facility for Industrial Transmutation (EFIT) prototype design with top level design parameters as delivered in the framework of the EUROTRANS project [5].

The parameters used for this facility and its fuel cycle are summarised in Table 1.

Table 1: Top-level EFIT design parameters

Target k_{eff}	0.97 (BOC)
Core inventory	5 325 kgIHM
Thermal power	384 MW_{th}
Discharge burn-up	78.28 MWd/kg
Fuel management	3 batches/core
Cycle time	368 days

The assumed EFIT availability was 87%. To obtain high MA transmutation rates uranium-free oxide fuel has been utilised. The Pu and MA oxide have been inserted in a MgO inert matrix. The fuel composition considered in the present study was fixed with a 55% fraction of MA and 45% Pu. The reference transmutation strategy adopted in this kind of reactor was that of "42-0", by which 40.17 kg of MA are burned per every TWh_{th}, versus only 1.74 kg of Pu. This means that the EFIT reactor is neither a Pu burner, nor a Pu breeder [5].

Scenario 1: Detailed results

In order to stabilise the MA main stock inventory of both Groups A and B, the optimal number of ADS to be deployed according to Scenario 1 was investigated. It was found that in total 25 ADS EFIT (384 MW_{th}) units are required to achieve this goal. The ADS deployment pace is given below (see Figure 1).

The spent fuel inventory in interim storage of Group A is reduced to zero by reprocessing before the end of the century (by 2072), see Figure 2.

The total mass of spent fuel in interim storage for Group A remains constant until 2022, when the German spent fuel legacy is added. Finally in 2040 the reprocessing plants start their operation and reprocess all the spent fuel by 2072.

Minor actinide inventory

According to Scenario 1, in 2100 the minor actinide total inventory (Figure 3) will be approximately 267 tonnes, of which: *i)* americium: 185 tonnes; *ii)* neptunium: 63 tonnes; *iii)* curium: 19 tonnes.

Two MA inventories are compared in Figure 4: *i)* build-up in the regional approach; *ii)* accumulated in Group A and B together in case no P&T strategy is deployed (*i.e.* no ADS units deployed).

The advantage of a regional scenario is clear: in 2100 the percentage difference of MA inventories is equal to about 40%, and in 2200 up to 104%.

Figure 1: ADS electric power production vs. time

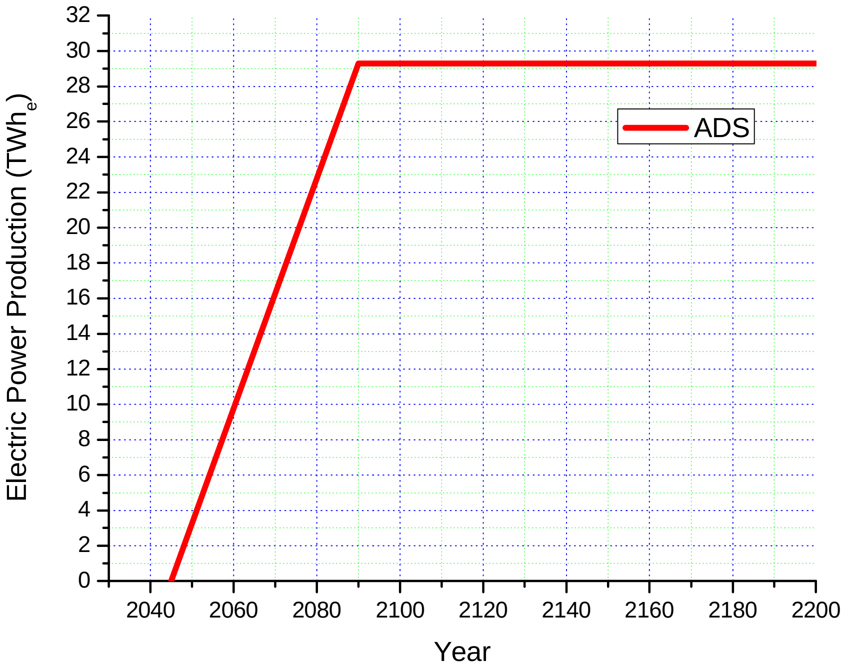


Figure 2: Spent fuel cumulative inventory (in tonnes) in Group A spent fuel interim storage

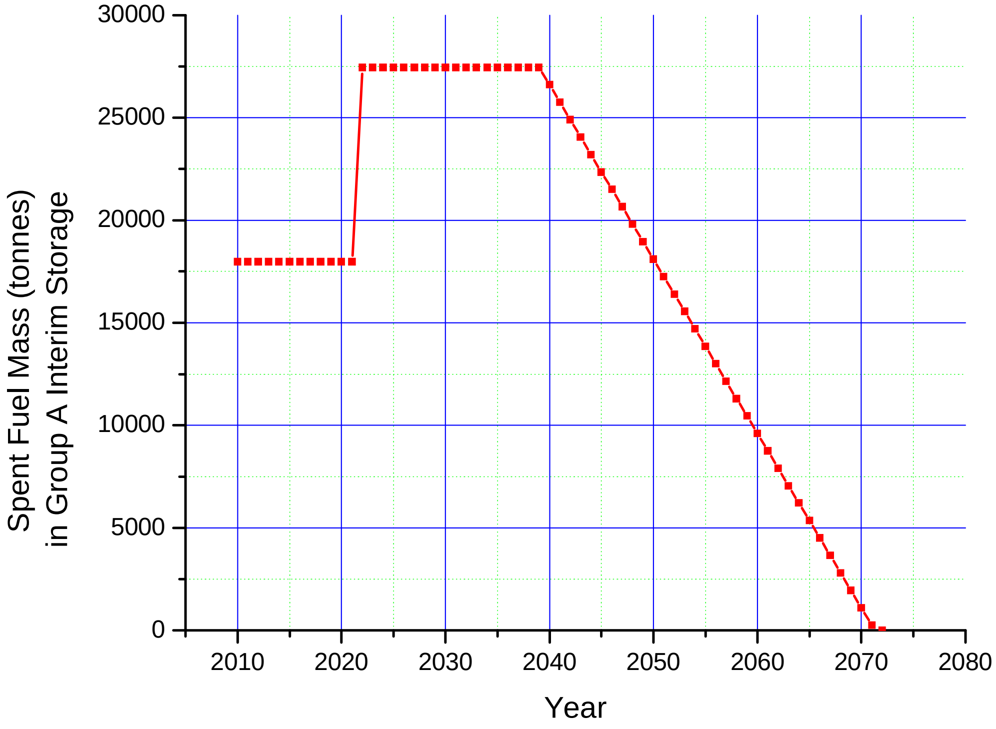


Figure 3: Minor actinide total mass (in tonnes) in all facilities

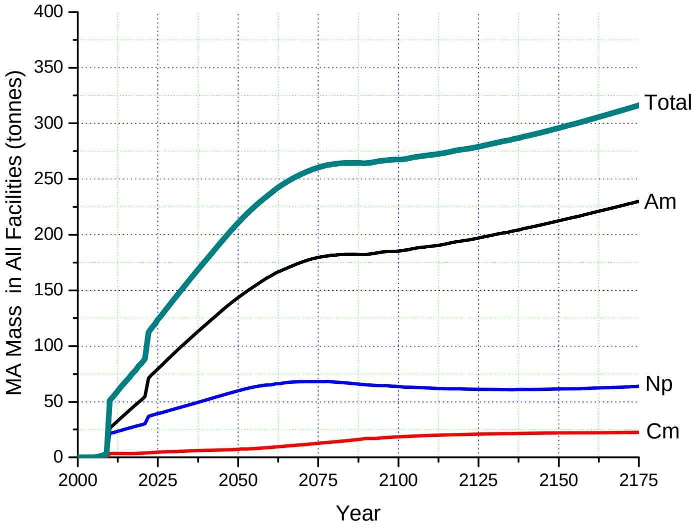
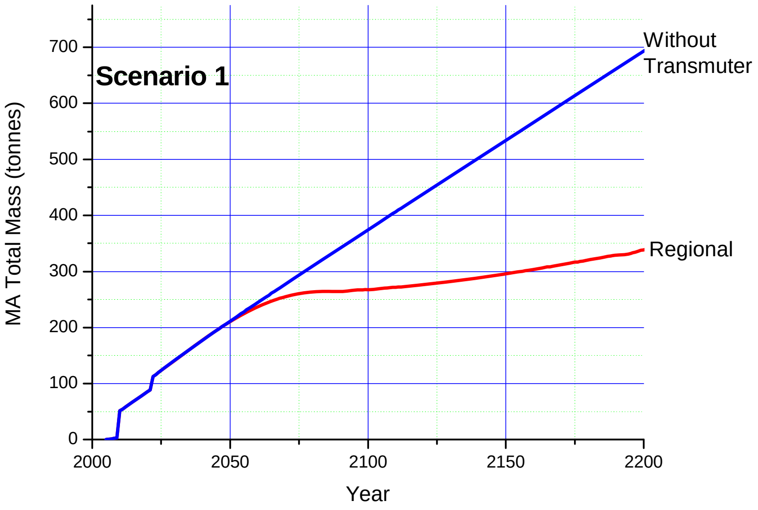


Figure 4: Total MA mass comparison between regional (Scenario 1) and “no transmuter” case



Scenario 1: Required facilities

Fuel fabrication

It could be observed, that a capacity of ca. 1 000 t/y for UOX plants, 100 for MOX, and 30 for ADS is required, in accordance with the defined annual electricity production per fuel type (387 TWhe for UOX and 43 TWhe for MOX).

Fuel reprocessing

The reprocessing capacities required in Scenario 1 for the regional approach and the scenario without transmuter (*i.e.* no P&T) are described in Table 2.

The lesser reprocessing capacity in case of separate (without transmuter) scenario is given by the absence of Group A – phasing-out nations – (850 t/y) and ADS (850 t/y) reprocessing facilities: the difference between the two cases considered in the table is in fact 1 700 t/y (half of which, as stressed in the text, of “new type”, *i.e.* ADS fuel)

Table 2: Reprocessing capacities (in tonnes/year) for regional (Scenario 1) and "no transmuter" approach

Scenario 1			
Regional		Without transmuter (no P&T)	
UOX	2 350	UOX	1 500
MOX	500	MOX	500
ADS	850	ADS	0
Total	3 700	Total	2 000

Scenario 2: Detailed results

In order to assess and stabilise the stockpile of separated MA arising from the spent fuel reprocessing of both Group A and B, the optimal number of ADS to be deployed was investigated. It was found that in total 27 ADS of EFIT type are required to achieve this goal (two more units than with respect to Scenario 1).

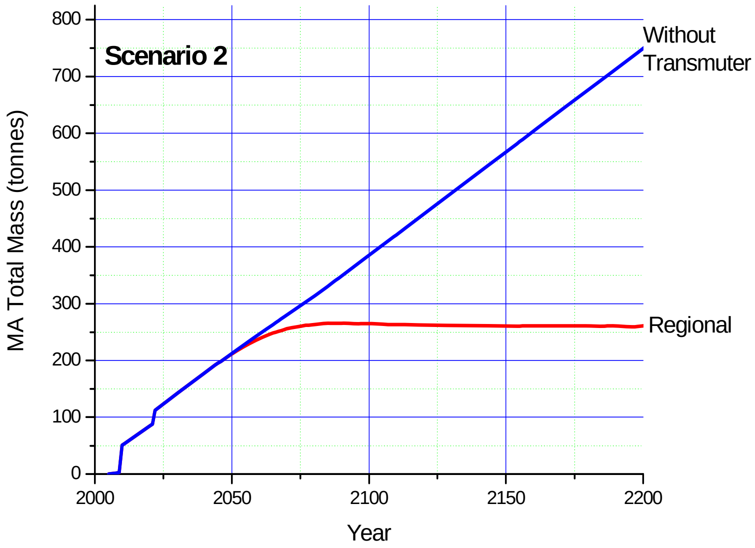
The ADS energy production is shown in Table 3:

Table 3: Deployment pace of ADS fleet (energy demand)

Time period	Scenario 1	Scenario 1	Scenario 2	Scenario 2
	Annual energy production (TWh_e)	No. of total ADS deployed	Annual energy production (TWh_e)	No. of total ADS deployed
2045	0	0	0	0
2090-2200	29.3	25	31.6	27

In Figure 5 two MA inventories are compared: *a)* accumulated in the regional approach, and *b)* build-up in Group A and B together without transmuter (*i.e.* no P&T: no ADS units deployed) approach.

Figure 5: Total MA mass comparison between regional (Scenario 2) and "no transmuter" case



The advantage of a regional scenario is clear: in 2100 the percentage difference of MA inventories is equal to about 45%, in 2200 up to 187%.

Scenario 2: Required facilities

Fuel fabrication

As for Group B, the annual fabrication capacity of about 690 t/y for UOX-fuelled plants, 90 for MOX and 300 for MOX RMA is required. The fabrication capacity needed for ADS is approximately 40 tonnes per year.

Fuel reprocessing

The reprocessing capacities required in Scenario 2 for the regional approach as compared to the case without transmuter (*i.e.* no P&T) are described in Table 4.

Here again, the differences are caused by the absence of ADS and associated reprocessing facilities.

Table 4: Reprocessing capacities for regional (Scenario 2) and "no transmuter" approach

Scenario 2			
Regional		Without transmuter (no P&T)	
UOX/MOX	2 450	UOX/MOX	1 600
ADS	850	ADS	0
Total	3 300	Total	1 600

Comparison between regional Scenarios 1 and 2 and the case without P&T

In order to keep the out-of-pile fuel inventory in interim storages as low as possible (with a given cooling time) both in Scenarios 1 and 2, and in order to reduce the consequent middle- and long-term radiotoxic impact on repository, the number of required ADS units to be deployed is much higher than the number required just to stabilise the MA inventory. For Scenario 1, a total of 25 units were necessary to achieve this goal (vs. 16 in order to stabilise only the MA main stock inventory); whereas 27 units are needed for Scenario 2.

If only the MA stabilisation goal is pursued, the advantage of the regional approach is evident from the COSI6 simulations: a smaller number of ADS units have to be employed (or, otherwise, with the same number of transmuters the MA inventory is not simply stabilised, but is reduced down to 0).

An important point is the reduced need of separate fuel cycle installation while comparing the regional deployment of P&T and the deployment of P&T by countries "in isolation". In fact, in the regional case there is no need for specific installations in Group A.

Due to the specific EFIT performance, which does not burn plutonium, this type of transmuter is suitable essentially for a regional approach, where the excess amount of plutonium is used to generate more energy. However this feature is a serious drawback when considering nations phasing-out nuclear "in isolation", as will be discussed below.

Finally, the particular composition of ADS EFIT fuel should be considered (45% Pu, 55% MA), since it would require specific technology developments (particularly complex, considering that the reprocessing and fabrication of ADS fuel is not yet a proven technology).

A point to be taken into account in the mono-recycling scenario is the ageing of the Pu in FR stock. In fact a considerable amount of ^{241}Am will be generated by radioactive decay over time, forcing to shorten the storage period as much as possible by a more aggressive fast reactor fleet start-up.

Concerning the Pu multi-recycling option (*i.e.* with recovery of plutonium from spent fuel) it allows extracting more energy from the mined uranium, thus slightly enhancing the fuel, and consequently, natural resource utilisation. The multi-recycling of Pu in the LWR induces a significant increase of MA production, compared to the mono-recycling case. However, this contribution is less important than the difference in the Pu vector. The drawback is finally a lesser (perhaps insufficient) quantity of plutonium available for a future fast reactor fleet.

If Group A pursues independently the P&T transmutation strategy, the ADS units should obtain the plutonium needed for their start-up cores from its (*i.e.* Group A) spent nuclear fuel. In 2022 the Group A spent fuel legacy should contain, according to a preliminary assessment, ~320 tonnes of plutonium and ~44 tonnes of MA. Since plutonium constitutes 88% of the TRU mass, the ADS used to transmute that TRU must necessarily employ relatively short cycles. It was shown in Ref. [6] that the steep burn-up reactivity gradient resulting from use of Group A TRU inventory limits the ADS cycle burn-up to 40 MWd/kg (with a reactivity swing $\Delta k_{eff} = 0.03$) and the cycle time to slightly less than one half of a year (168 days), in case of a sodium-cooled metal-fuelled ADS (840 MW_{th} and LBE target). The very low Pu transmutation rate of EFIT (less than 2 kg/TWh_{th}) would imply for phasing-out nations of Group A no significant reduction in total Pu inventory and the lack of benefit which the destruction of Pu isotopes (especially ^{238}Pu) brings for disposal options. These restrictions of ADS performance constitute a drawback for Group A acting alone and might daunt the nations phasing-out from choosing EFIT as dedicated facility to transmute their TRU.

Parametric studies on Scenario 3

A major objective of this scenario is the consumption of the TRU legacy stocks of Group A at a reasonable time horizon. It was shown in Ref. [4] that there exist a number of options in that respect, since the time necessary for the consumption of the TRU of Group A depends on a number of parameters, such as:

- *The breeding gain of the FR in Group B*: the lower the gain, the more efficient the FR for transmutation (the time necessary for the consumption of the actinide inventory can be reduced by using the CAPRA concept [7] in the transition period).
- *The out-of-reactor time in Group B*: the longer this time, the more TRU needed from Group A for the fuel cycle TRU inventory in Group B.

It was shown in Ref. [4] that, whatever the out-of-reactor time, with a FR having a low breeding gain (-0.196), the TRU inventory of Group A is consumed within less than 100 years. With 1+2 years and an intermediate breeding gain (-0.061), there is almost no impact on TRU inventory of Group A.

The other important parameters are:

- number of countries involved in the regional approach (impact of the initial inventories);
- date and pace of deployment of the fast reactors;
- load factor in PWR and FR (more electricity can be produced with the same installed capacity);
- production of Pu in the PWR which can be increased if necessary by reducing the burn-up of the fuel;
- preliminary step for Pu as a MOX in PWR.

Figure 6 summarises the trends of the dependence of the time needed to consume the TRU of Group A countries on both parameters. Later in this work, the results of the scenario corresponding to the low breeding gain (-0.196) and one year of cooling will be presented in detail, because these assumptions allow to:

- have the shortest time for TRU consumption in Group A;
- deploy fast reactors in Group B without lack of TRU.

Parametric studies on Scenario 4

An important outcome of this scenario is the potential for restarting a sizable nuclear power fleet in some countries of Group A. Here again, a parametric study was performed in order to indicate significant trends and options and the main results have been described in Ref. [4] and are shown in Figure 7.

Figure 6: Scenario 3 – parametric studies

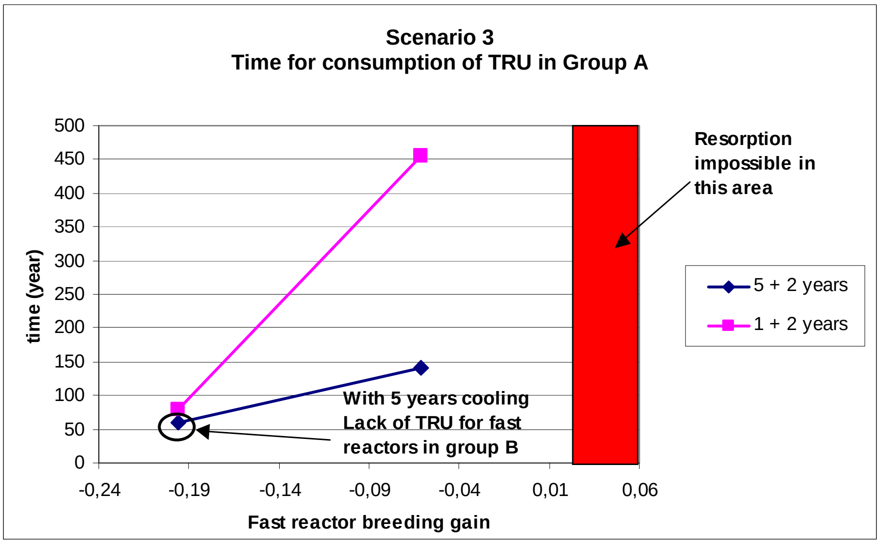
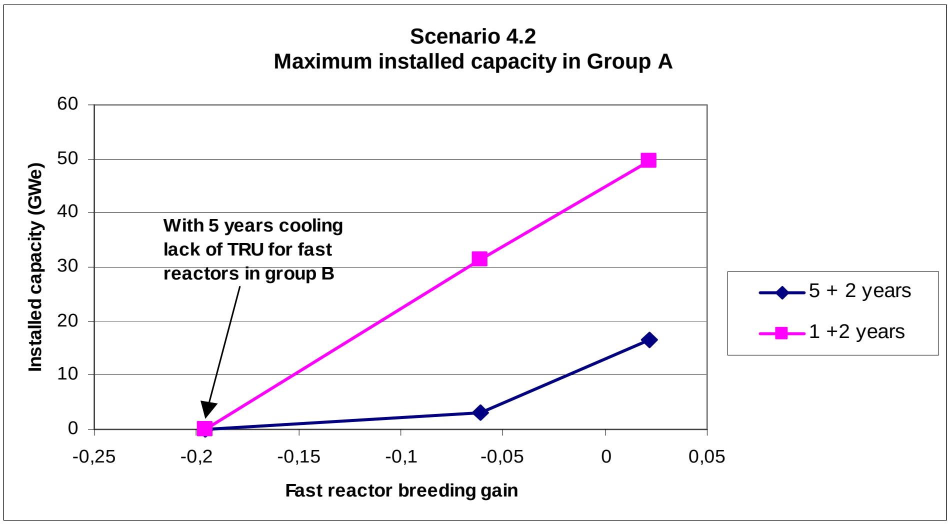


Figure 7: Scenario 4 – parametric studies



The parametric study did show that the maximum installed capacity in Group A depends on:

- the breeding gain of the fast reactors (the higher the breeding gain, the more reactors can be deployed);
- out-of-reactor time for TRU, which determines the TRU fuel cycle inventory.

With an out-of-reactor time of 1+2 years for a fast reactor fuel cycle, the deployment of new fast reactors in Group A is still possible in 2050.

The maximum achievable installed capacity corresponds to 49.5 GWe in Group A, corresponding to 330 TWhe (load factor = 76%, breeding gain = +0.022). The pace of deployment is 1.5 GWe/year.

An out-of-reactor time of 5+2 years would allow a lower achievable installed capacity.

As in the previous case, other important parameters can play a significant role in the scenario overall assessment:

- number of countries involved in the regional approach (impact of the initial inventories);
- date and pace of deployment of the fast reactors in both groups;
- date and pace of decommissioning of the PWR in Group B;
- load factor in the PWR and FR;
- breeding gain in the fast reactors which can be fitted to produce or to adjust the required Pu inventory;
- production of Pu in the PWR which can be increased if necessary by reducing the burn-up of the fuel.

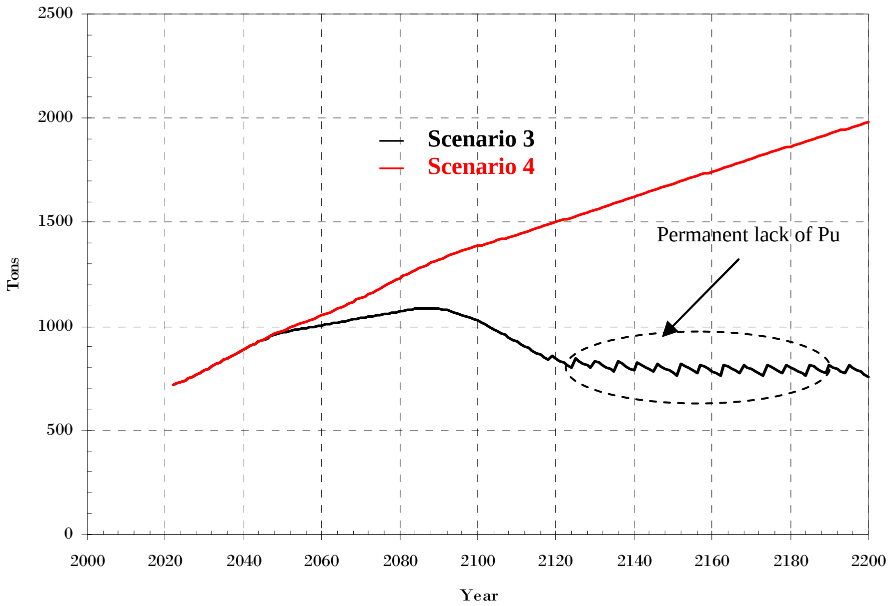
Later in this work, the results of the scenario corresponding to the higher breeding gain (+0.022) and 1 year of cooling will be presented in detail, as this allows deploying the maximum installed capacity in Group C.

Detailed results of Scenarios 3 and 4

Plutonium inventory

Figure 8 gives the plutonium inventories in the cycle for Scenarios 3 and 4.

Figure 8: Scenarios 3 and 4 – plutonium inventory in the cycle



For Scenarios 3 and 4, the evolution of the Pu inventory depends on the breeding gain of the fast reactors.

For Scenario 3, the objective was to eliminate as quickly as the possible the TRU inventory of Group A and the FR chosen have a negative breeding gain. In this case, the total Pu inventory decreases once the entire fleet of FR are deployed.

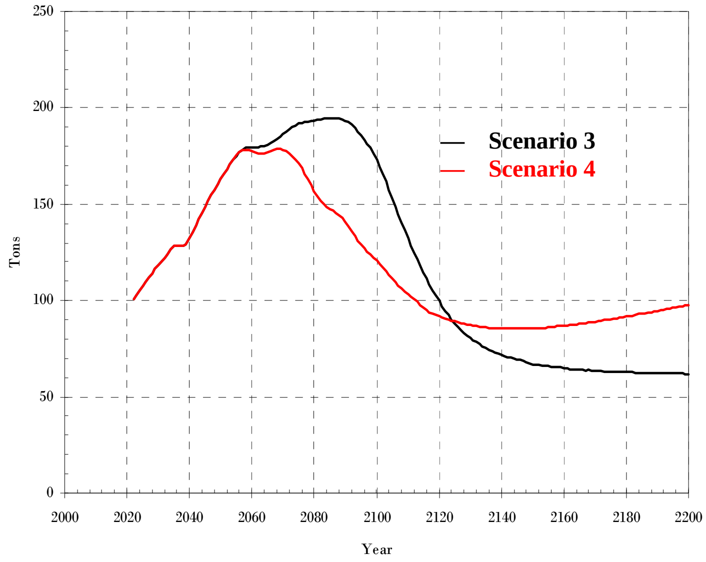
Due to negative breeding gain, a permanent lack of fissile materials appears from the year 2120; lacking Pu is provided by an external Pu source ("infinite Pu") and the inventory is artificially stabilised.

For Scenario 4, the objective was to restart as many FR as possible in Group C. That is the reason why FR with positive breeding gain has been chosen. In this case, the Pu inventory increases.

Minor actinide inventory

Figure 9 gives the minor actinide inventories in the cycle for Scenarios 3 and 4.

Figure 9: Scenarios 3 and 4 – minor actinide inventory in the cycle



For Scenarios 3 and 4, the evolution of the MA inventories depends on the transmutation strategy applied. In both scenarios, the MA are transmuted in the FR as soon as they are deployed. For that reason, the total MA inventory decreases when enough FR are deployed.

At the end of Scenario 4, the MA inventory increases because of the positive breeding gain.

Scenarios 3 and 4: Required regional facilities

For Scenario 3, the regional facility to be deployed is the reprocessing plant (or plants) to be used by both Group A and Group B countries. For Group A, the reprocessing capacity needed depends on the time of consumption of TRU. Considering the total amount of spent fuel in Group A, *i.e.* 29 000 tonnes (mainly UOX) and the minimum time for reprocessing of this fuel, *i.e.* 35 years, the maximum reprocessing capacity for the needs of Group A is 850 tonnes of initial heavy metal per year. This value can be reduced by increasing the period for reprocessing. For Group B, an additional capacity of 850 tonnes per year for PWR UOX and MOX spent fuel is sufficient for the deployment of fast reactors. An additional capacity of 300 tonnes in order to reprocess FR spent fuel is necessary at the end of the century (100% fissile fuel).

For Scenario 4, the regional facilities needed are the reprocessing plant for Group A and Group B and the fuel fabrication plant for the fast reactors of Group A and B.

Reprocessing facility

For Group A, the maximum reprocessing capacity is necessary at two different stages. At first, for the reprocessing of the initial PWR spent fuel, *i.e.* approximately 850 tonnes per year (as for Scenario 3). Successively, when fast reactors are deployed, the maximum installed capacity in Group A related to the newly deployed fast reactors is 49.5 GWe (GRG = 0.022 and one year of cooling). The reprocessing capacity is then approximately 400 tonnes per year (61% of fissile fuel and 39% of blankets). For Group B, the required reprocessing capacity is around 500 tonnes per year (61% of fissile fuel and 39% of blankets). This value is higher than for Scenario 3 because the fast reactors also contain axial and radial blankets which need to be reprocessed.

Fabrication facility

The full fabrication capacity for the fast reactors of Groups A and B is needed at the end of the century, when all fast reactors are deployed.

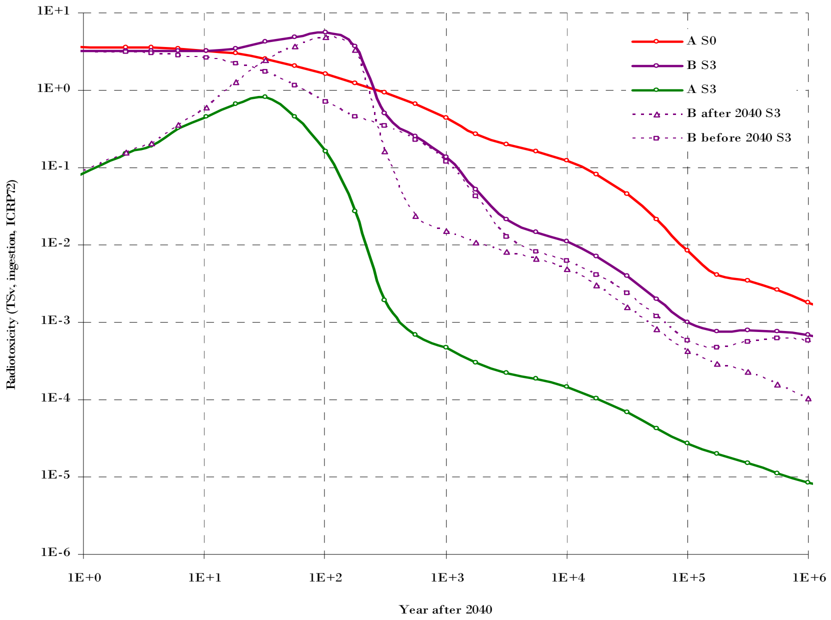
As for Group A, since the maximum installed capacity is 49.5 GWe (GRG = 0.022, one year of cooling), the annual fabrication capacity should be approximately 400 tonnes per year (61% of fissile fuel and 39% of blankets).

As for Group B, since the maximum installed capacity is 64.5 GWe at the end of the century (430 TWhe per year, load factor = 0.76%), the fabrication capacity needed is approximately 500 tonnes per year (61% of fissile fuel and 39% of blankets).

Impact on waste disposed

Figure 10 gives the cumulated radiotoxic inventory of the HLW in Groups A and B for Scenario 3. The HLW accounted are the waste produced between 2010 and 2200. The radiotoxic inventory is presented from 2040, date of the beginning of the partitioning of the MA. The increase of the "B S3" plots (total value given by the solid line; contributions before and after 2040 given by the dashed lines) during the first 100 years is due to the HLW produced by the reprocessing of fast reactor spent fuel and blankets.

Figure 10: Scenario 3 – radiotoxic inventory of the high-level waste



The red plot gives the evolution of the radiotoxic inventory of the spent fuel of Group A in case of no reprocessing and transmutation strategy is applied for TRU. The green plot indicates that the radiotoxic inventory of the HLW has been reduced by a factor 1 000. The reduction is due to the separation efficiency of the regional reprocessing plant (0.1% for U, Pu and MA). The plot "B before 2040" indicates that the major part of the radiotoxic inventory in Group B is coming from the HLW produced before P&T implementation. It should be noted that the initial HLW in 2010 have not been accounted for in these calculations.

Figure 11 gives the decay heat of the cumulated HLW in Groups A and B for Scenario 3. The HLW accounted for are the waste produced between 2010 and 2200.

The red plot (A S0) gives the evolution of the decay heat of the spent fuel of Group A in case of no reprocessing and transmutation strategy is applied for TRU. The green plot (A S3) indicates that the decay heat of the HLW after 100 years of cooling has been reduced by a factor 10. At this time, the contribution of the fission products to the decay heat of the HLW remains important.

Figure 11: Scenario 3 – decay heat of the high-level waste

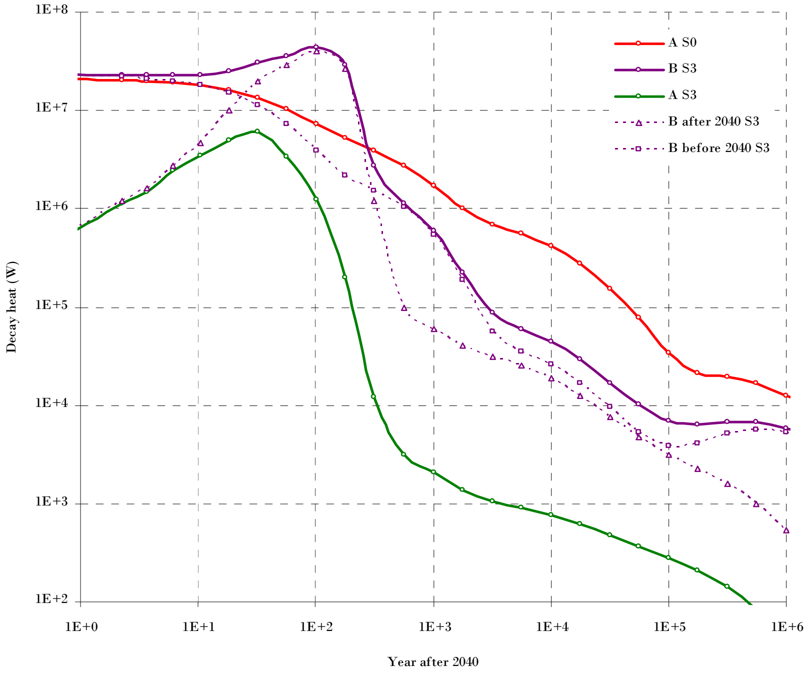
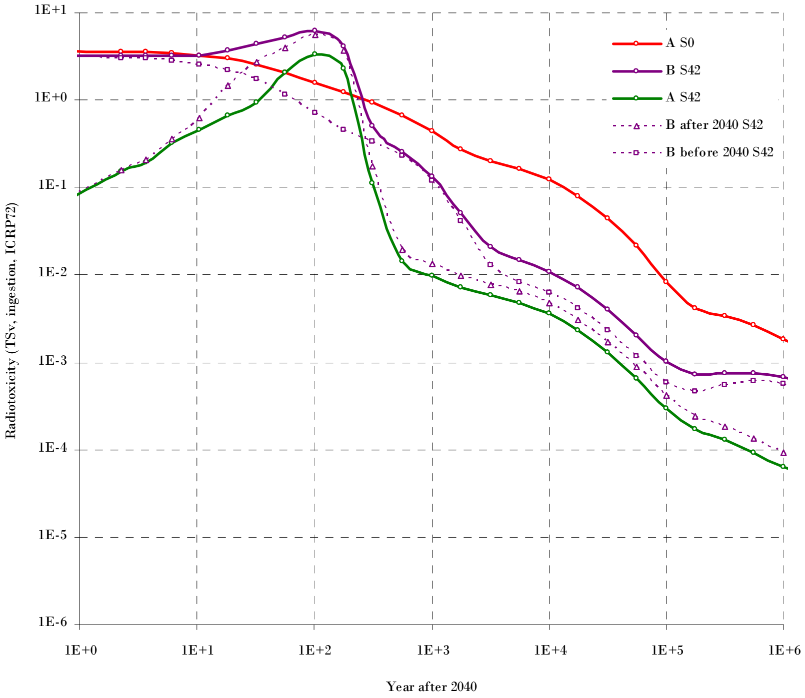


Figure 12 gives the cumulated radiotoxic inventory of the HLW in Groups A and B for Scenario 4. The HLW accounted for are the waste produced between 2010 and 2200.

Figure 12: Scenario 4 – radiotoxic inventory of the high-level waste



Result for Group B

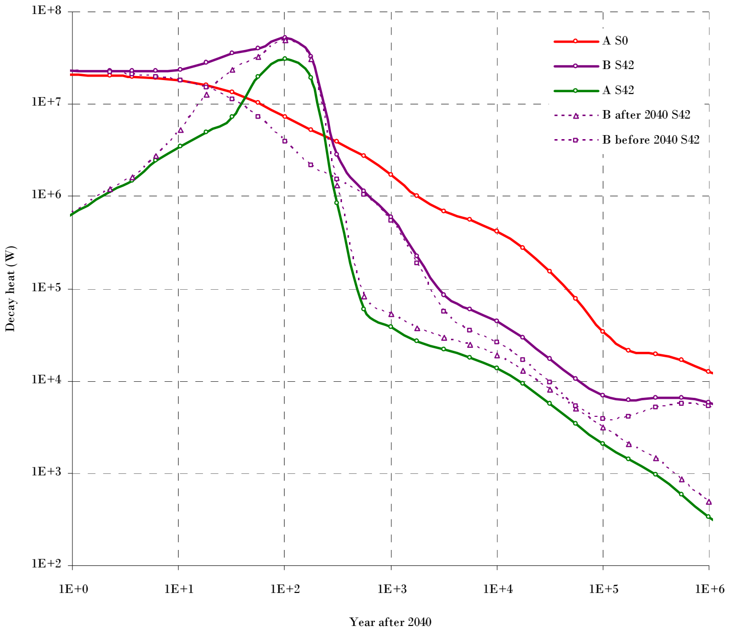
For a cooling time higher than 500 years, the radiotoxic inventory is dominated by the waste produced before the deployment of P&T, as for Scenario 3.

Result for Group A

For a cooling time higher than 500 years, the radiotoxic inventory is decreased by a factor higher than 10, compared to the direct disposal of spent fuel. It should be noted that the annual electrical production in this group is 330 TWhe, compared to the direct disposal case where no electricity is produced.

Figure 13 gives the decay heat of the cumulated HLW in Groups A and B for Scenario 4. The HLW accounted for are the waste produced between 2010 and 2200. The same trends are obtained as for radiotoxic inventory.

Figure 13: Scenario 4 – decay heat of the high-level waste



Summary on Scenarios 1 and 2

The main results of the regional P&T Scenarios 1 and 2 can be summarised as follows.

The spent fuel stock of Group A can be decreased, as required, down to 0 by 2100: all the fuel was reprocessed by that date. In Scenario 1 the Pu mono-recycle option implies that the reprocessed plutonium is kept in order to be successively transferred to fast reactor plutonium stock; it will be introduced in a later stage in FR fuel cycle of Group B. In order to stabilise the MA production from Group B, the required number of ADS of the EFIT type (384 MW_{th}) was determined to be 25 units for Scenario 1, and 27 for Scenario 2 (due to plutonium multi-recycling and consequently higher MA generation). The final plutonium inventory available in Scenario 1 at 2100 for the fast reactor fleet will be 840 tonnes. The results of the Scenario 2 simulation with COSI6 show that the plutonium main stock inventory of Group B is stabilised starting from 2100 at ca. 100 tonnes, while the total inventory increases slightly vs. time due to accumulation of "bad quality" plutonium coming from the MOX multi-recycling (which is associated however to a lesser MA production by radioactive decay with respect to plutonium mono-recycling).

For the proposed transmutation strategy, a total reprocessing capacity of 3 700 tonnes per year is needed in the case of Scenario 1 and of 3 300 tonnes in the case of Scenario 2: 850 tonnes per year for

ADS reprocessing plant, 850 for Group A spent fuel legacy, and 2 000 for Group B in Scenario 1, 1 600 for Group B in Scenario 2. The PWR fuel reprocessing capacity required for Scenario 1 is about 18% higher than that currently available in France, while the ADS reprocessing facilities must obviously be developed and deployed in the future.

In Scenario 1 the annual capacity of fuel fabrication plants should be: 1 000 tonnes/year for UOX, 100 for MOX and 30 for ADS. In Scenario 2 the needed capacity is: 690 tonnes/year for UOX, 390 for MOX and 40 for ADS. The total capacity is quite similar in the two cases considered, while only MOX/UOX fabrication capacities proportions are sensibly different (respectively, 1:10 and 1: 1.77).

As a final consideration on the EFIT design, we should mention that this type of transmuter offers potential benefits only in regional scenarios, but does not appear suitable for phasing-out nations implementing a P&T strategy in isolation, because its transmutation performance was focused exclusively on MA and leaves most of the Pu stock unchanged.

Summary on Scenarios 3 and 4

The major result of Scenarios 3 and 4 is the possibility to manage the Pu and MA of several countries through a regional approach with fast reactors either burners or breeders, depending on the strategies applied in the countries.

It could be important to use the flexibility of a fast reactor and the reversibility from burner to breeder, as demonstrated in past studies (see *e.g.* Ref. [8]). This would allow making the transition from burner to breeder configurations for the fast reactors (all or part of them) at a specific point in time. This feature can allow tuning future strategies at a regional level that account both for sustainability and for waste minimisation.

A significant reduction of the radiotoxic inventory of the high-level waste is obtained for both groups of countries, even in the case of a restart of nuclear energy in Group A.

Moreover, the added value of the fast reactors compared to the ADS is the electricity produced (330 TWhe per year in Group A in Scenario 4).

It has also been pointed out that the optimisation of scenarios depends, as expected, on a number of parameters, and, among them, parameters that characterise the fuel cycle (cooling times, etc.) are particularly significant and will have to be investigated in detail.

Overall summary and perspectives

Regional strategies can in principle provide a framework for implementation of innovative fuel cycles, with appropriate sharing of efforts, accounting for proliferation concerns and resource optimisation. In the present paper we have presented the first results of specific scenarios, presently investigated within a wider effort under way in Europe in order to shape a roadmap to implement partitioning and transmutation technologies.

The indications obtained so far underline that if, *e.g.* fast reactors with homogeneous recycle of non-separated TRU are envisaged, there is a need to optimise the fast reactor characteristics (*e.g.* the conversion ratio), and the fuel cycle characteristics (*e.g.* the fuel out-of-pile cooling time), in order to meet the potentially different objectives of different countries within a regional area.

In the present study, we did not investigate the impact of introducing critical fast reactors using heterogeneous recycle of MA, and this can be the object of future studies. However, the potential limitations in terms of maximum allowed amount of MA that can be loaded in a target and the potential absence of fertile blankets can reduce the flexibility of fast reactors, as discussed above, which allow coping with a range of objectives within a regional area.

Another relevant finding of the study is related to the characteristics of the ADS chosen to transmute MA in scenarios of the “double strata” type. In fact, most ADS design studies, and in particular those performed in the framework of EUROTRANS, have considered a fuel loading and a transmutation potential mostly adapted to “MA”, and not “TRU”, consumption. Then, this type of ADS is more apt to be used in a “regional” scenario where different countries with different objectives do share resources, facilities and spent fuel inventories in order to minimise wastes. The same type of ADS will not be useful in the case of a country committed to a stagnant or decreasing use of nuclear energy that would decide to deploy P&T in "isolation" for waste management.

In this respect, an interesting addition to the present study would be the introduction of a critical "burner" fast reactor (*i.e.* with a conversion ratio in the range 0.5-0.8) in Scenarios 1 and 2.

As far as the impact of the implementation of P&T at a regional level, the results of the scenario studies indicate that the expected beneficial potential of P&T, *i.e.* reduction of the radiotoxicity in a repository to the level of the radiotoxicity of the initial ore after few hundred years, and the reduction of the heat load in the repository (more than one order of magnitude), applies to the whole region, providing a potential significant benefit to all the countries of that region (*e.g.* Europe), despite their different policies in terms of nuclear energy. Moreover, the present studies have shown the potential of a regional strategy in favour of a nuclear "renaissance" in some countries.

Further studies will obviously be needed, in particular in order to investigate practical issues (like fuel transport, etc.) and institutional issues which will, without a doubt, be very challenging.

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Global scenarios for fast reactor deployment*

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Abstract

We raise expectations for nuclear power development in Asia due to high economic growth rate and rush for energy demands. On the other hand, the movement of review of the nuclear energy role is spreading in developed countries against the backdrop of increases in the cost of fossil fuels and strengthening of reduction of greenhouse gases emissions. There are 429 nuclear power plants in operation with a capacity of 387 GWe in the world as of the end of 2006 and they are expected to increase from 4 to 40 times at the end of this century according to IPCC and IIASA reports. Though this widespread introduction of nuclear energy is projected to lead to global tight uranium supply and uranium resource depletion, fast reactor (FR) cycle is one of most realistic options for the realisation of sustainable nuclear energy. Even if the IIASA-C2 case is studied, in which nuclear electricity demand projection is relatively lower than other cases, the accumulative uranium demand in LWR once-through scenario will exceed the total conventional natural uranium resources at the end of this century. Therefore, FR need to be deployed commercially by 2050 at the latest in IIASA-C2 case in order to reduce the accumulative uranium demands under the conventional natural uranium resources limit. In FR deployment scenario, sufficient reprocessing capacity for spent fuels of not only LWR but also FR is needed to maintain plutonium supply and demand balance.

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* The full text of this paper was unavailable at the time of publication.

Scenarios for the deployment of sodium-cooled fast reactors in France

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Abstract

In the frame of the French law for the waste management, CEA has studied different solutions (fuel and reactor concepts) to manage plutonium and minor actinides in the French fleet of reactors. The insertion of a new concept must be evaluated in the global electronuclear system taking into account the whole fuel cycle (enrichment, fuel fabrication, reactors, interim storage, processing, waste storage). Each option has been evaluated in different dynamic scenarios from the present situation to the future generation systems (Gen-IV) [1]. The reference option for the Gen-IV reactors in France is the sodium-cooled fast reactor (SFR).

The rhythm of development and the level of electrical production achievable with fast spectrum systems depends on plutonium resources and many other assumptions as reactor design and fuel cycle assumptions. In a first part, this paper presents the impact of some reactor and fuel cycle assumptions on the possibility to deploy the SFR in France, from the plutonium inventory point of view.

The transmutation of minor actinides in the Gen-IV fast reactors can be obtained in two different modes. In the homogeneous mode, the minor actinides to be transmuted are directly mixed with the "standard" fuel of the reactor. In the heterogeneous way, the minor actinides are separated from the fuel itself, in limited the number of S/A (targets) devoted to minor actinides transmutation. This recycling offers the possibility to load a significantly high mass of minor actinides in the reactor and to separate (U, Pu) fuel fabrication with from minor actinides blanket fabrication.

The last neutronic studies for minor actinides transmutation in sodium fast reactor depleted uranium radial blanket are presented in [2]. The concept is based on an heterogeneous multiple recycling model (whole or part of minor actinides and plutonium at the end of each reactor cycle is sent back in the following cycle; only reprocessing losses go to the waste). The use of the oxide matrix allows to reprocess such S/A in the spent fuel standard flow. In a second part, this paper will present the impact of some transmutation assumptions on the plutonium and minor actinides management.

Introduction

Once the reactor transmutation performances of a new concept have been assessed, the insertion of this concept must be evaluated in the global electronuclear system with an analysis of the impact on the fuel cycle (enrichment, fuel fabrication, reactor, processing, interim storage, waste storage). An approach of scenario studies [3] is used to evaluate two scenarios in the French context:

- *Scenario 1*: Deployment of SFR recycling plutonium, minor actinides being sent to the waste.
- *Scenario 2*: Deployment of SFR recycling Pu in the fissile part of the core and minor actinides in the radial blankets.

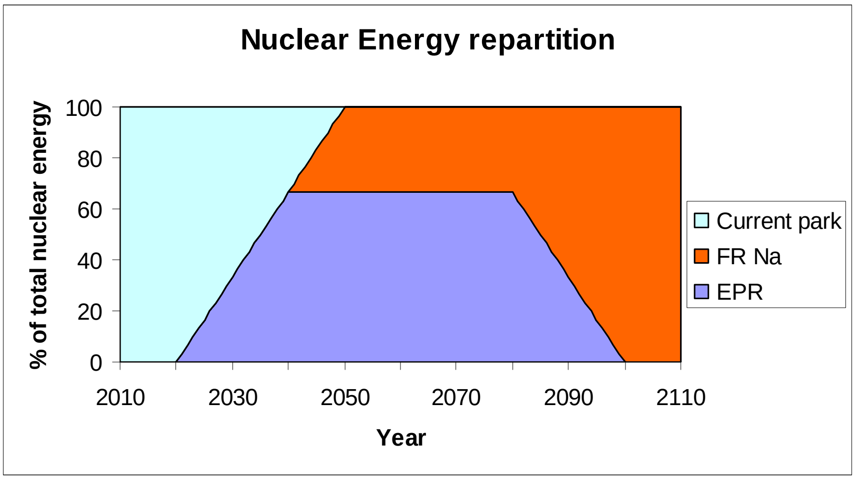
Both scenario consider the transition from the present situation in France (partially closed cycle) until the total closed cycle with Gen-IV systems [2].

This approach allows to calculate the fluxes and the inventories of these elements in each step of the fuel cycle and to treat the transient period to pass from one situation to the other. This study is a theoretical exercise, carried out by the CEA. It does not predict any underlying industrial strategy, the relevance of which would need to be assessed, according to industrial realities in an economic context possibly prevailing in the era under consideration.

The scenario assumptions

Both scenarios consider the French nuclear park as having a constant nuclear energy demand at 430 TWhe/year. This value corresponds to the current annual nuclear electricity production in France. The current nuclear park is replaced between 2020 and 2050 by a mixed nuclear park: 67% of Generation III EPR reactors and 33% of Generation IV SFR. The lifetime of the EPR and the SFR is 60 years, which means that the first EPR are replaced from 2080. From 2080 to 2100, the EPR are replaced by SFR. In 2100, the park has 100% of SFR, corresponding to 60 GWe.

Figure 1: Scenarios 1 and 2 – nuclear energy repartition



The other main assumptions are:

- The concept of fast reactor chosen is the SFR with a breeding gain close to zero without fertile blankets, in accordance with the Gen-IV specifications. To achieve this objective, the fraction fuel is increased and the Na fraction is decreased compared with EFR core. Thus, loaded Pu fraction is reduced, making it possible to approach the isogeneration. With radial blankets (Scenario 2), the breeding gain becomes positive thanks to Pu production in the blankets.
- The date of deployment of the first SFR is 2040. At this date, it is considered that the SFR technology is mature.

- The load factor of EPR and SFR is 81.76%.
- The pace of introduction of the EPR and SFR is 2 GWe/year, the same as the shutdown pace of the current fleet.
- The replacement of the second park starts from 2080.
- The single recycling of Pu in the LWR stops in 2038.

Core design and reactor assumptions

The main neutronic data of the SFR core are displayed in the Table 1. Two types of core design are chosen: Version 0 developed in 2006, and the more advanced Version 2B developed in 2008.

The plutonium inventory in the SFR V2B concept is higher than in the SFR V0. For this reason, the plutonium margin for the deployment of the fleet will be lower.

The main reactor assumptions are displayed in Table 2.

Table 1: SFR – assembly and core characteristics

Assembly and core characteristics	V0	V2B
Number of fissile fuel assemblies	424	454
Heavy nuclide volume fraction (vol.%)[*]	47.4	43.7
Na volume fraction (vol.%)	27.1	27.6
Structure volume fraction (vol.%)	18.5	20
Average core power density (W/cm^3)[†]	231	206
Reloading frequency	5	5
Average BU (GWd/t)	106	98
Maximum BU (GWd/t)	184	148
Average Pu enrichment (vol.% fuel)	14.7	15.8
Equivalent ^{239}Pu enrichment mass (%)	10	10.96
Initial Pu mass (kg)[‡]	10 472	12 043
Total heavy nuclides mass (tonnes)	71	74
Average instantaneous breeding gain BOC/EOC	0.053	+0.015
Void effect ($) BOC/EOC (with plenum)	2.6/4.5	5.6/6.0
Doppler fuel 1227°C → 2700°C BOC/EOC ($)	-1.8/-1.4	-1.3/-1.3

[*] Fuel porosity = 0.96.

[†] Volume is calculated at T = 20°C.

[‡] Pu vector: ^{238}Pu = 3.57/^{239}Pu = 47.39/^{240}Pu = 29.66/^{241}Pu = 8.23/^{242}Pu = 10.38/^{241}Am = 0.78.

Table 2: Reactor assumptions

	EPR	SFR
Thermal power	4 500 MWth	3 600 MWth
Net electrical power	1 550 MWe	1 450 MWe
Load factor	81.76%	81.76%
Core management	4* 366.6 EFPD	5 * 410 EFPD
Net yield	34.44%	40.3%
Fuel assemblies average burn-up	55 GWd/tonnes	106 GWd/tonnes – SFR V0 98 GWd/tonnes – SFR V2B
Fuel type	UOX, 17 × 17 MOX pins	See Table 1

The fuel cycle assumptions

The fuel cycle assumptions are displayed in Table 3.

The standard MOX spent fuel is reprocessed from 2038, in dilution with UOX spent fuel, to feed the SFR with plutonium.

For the calculations, the tool used is the code COSI [4].

Table 3: Scenarios 1 and 2 – fuel cycle assumptions

Enrichment	
Depleted uranium enrichment	0.25% until 2019 0.2% from 2020
Fabrication	
Standard MOX and SFR fuel fabrication time	2 years
Reprocessing	
Minimum cooling time before reprocessing	PWR fuel: 5 years SFR fuel: 2 or 5 years
Reprocessing losses	Scenario 1: 0.1% for U, Pu Scenario 2: Until 2037: 0.1% for U, Pu, 100% for Am, Np, Cm From 2038: 0.1% for U, Pu, Am, Np, Cm
Priorities for reprocessing	First in – first out until 2037 First in – last out from 2038

Scenario 1: Recycling of Pu in the SFR, MA go to waste

The results of Scenario 1 are displayed in Table 3.

Table 3: Scenario 1 – results

	SFRV0	SFR V2B
SFR fuel cooling time = 2 years ^{239}Pu equivalent margin Installed capacity margin (GWe)	 +214 tonnes +27 GWe	 +90 tonnes +10 GWe
SFR fuel cooling time = 5 years ^{239}Pu equivalent margin Installed capacity margin (GWe)*	 +61 tonnes +6 GWe	 -105 tonnes[†] -9 GWe

* This margin takes into account the total necessary Pu mass: for reactors, fabrication plant and the Pu contained in the spent fuel cooling.

[†] The lack of Pu appears after 2090.

The plutonium margin depends on the SFR design and fuel cycle assumptions. The margin can be either positive, which means that we have the possibility to deploy more SFR; either negative, which means that an external source of plutonium is necessary to deploy 60 GWe of SFR in 2100. Since this design is not fixed today, it is interesting to perform some sensitivity studies, so as to assess the impact of the SFR design and fuel cycle assumptions on the Pu margin.

The parameters having an impact on the Pu margin are:

- Thermal power: 3 600 MWth (data).
- Volumic power: variable, depending on SFR design.
- Cycle length: 410 EFPD, depending on core design.
- Number of cycle: 5.
- Heavy nuclide mass: variable depending on SFR design.

- Maximum fuel assembly burn-up: 150 000 MWd/t (design criteria).
- Average fuel assembly burn-up: 100 000 MWd/t (hypothesis: form factor = 1.5).
- Minimum cooling time before reprocessing: variable.
- Fabrication time: 2 years.
- Equivalent ^{239}Pu mass: depends on 2, 3, 4, 5, 7.
- SFR net yield: 40.3%.

Most of these quantities are connected to each other. For example:

$$Burn-up\max = \frac{Thermal\ power * Form\ factor * Cycle\ length * Number\ of\ cycles}{Heavy\ nuclides\ mass}$$

Given the large number of parameters involved in the plutonium balance, it was decided to retain three parameters for the parametric studies:

- Two core parameters: Equivalent ^{239}Pu mass and mass of heavy nuclides in the core. The cycle length can be deduced from these two parameters with the help of average discharge burn-up.
- One cycle parameter: the minimum cooling time of SFR spent fuel before reprocessing.

For each configuration (Pu mass, heavy nuclides mass, minimum cooling time), a COSI calculation is made. Other calculation assumptions are the same for all the calculations. The results are presented in Figures 2 and 3, giving the margin, in terms of SFR installed capacity.

It is important to note that the plots do not represent a possible design for the SFR core. The aim of these graphics is to give some sensitivity effects of some SFR design data for the plutonium inventory necessary for the deployment of the SFR.

Figure 2: Scenario 1 – parametric studies – SFR spent fuel cooling time is two years

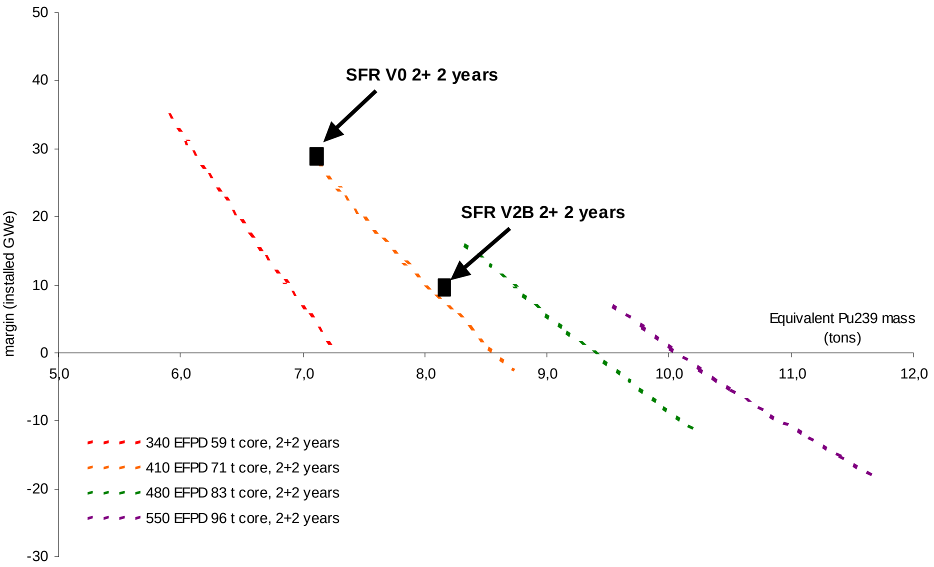
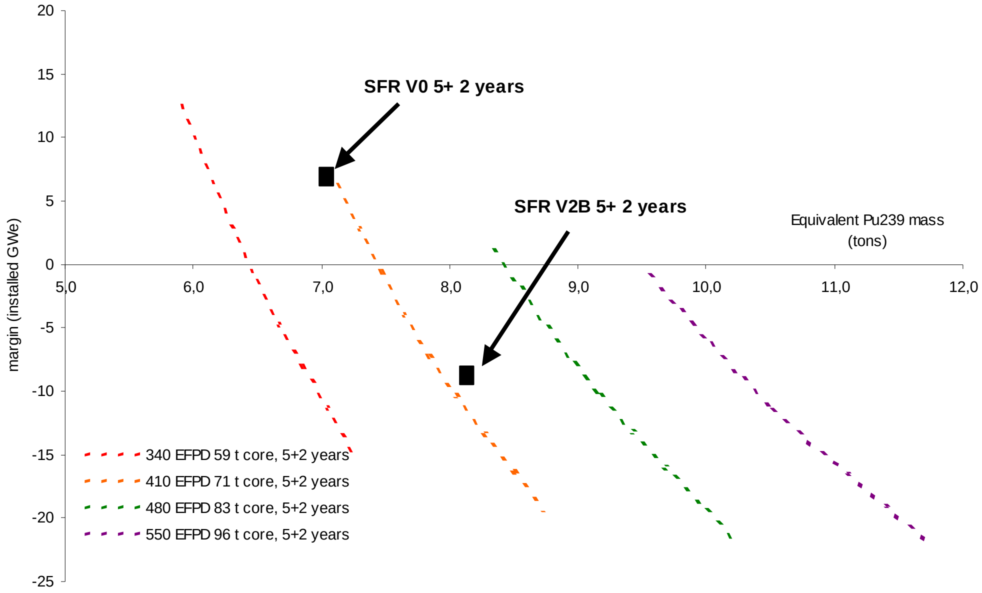


Figure 3: Scenario 1 – parametric studies – SFR spent fuel cooling time is five years



The results indicate that:

- The margin in terms of plutonium and installed capacity is very sensitive to the equivalent ^{239}Pu mass in the core. For example, an increase of 10% of the equivalent ^{239}Pu mass in the core induces a decrease from 9 GWe to 15 GWe for the possible installed capacity (no external source of plutonium is taken into account).
- There is no lack of Pu in the first stage of deployment of SFR (2040-2050).
- In case of lack of Pu, it appears at the end of the second stage of the deployment of the SFR, after 2090.

Scenario 2: Deployment of SFR recycling Pu in the fissile part of the core and minor actinides in the radial blankets

In Scenario 2, the minor actinides are recycled in the radial blankets of the SFR from 2040. One range of radial blankets is considered and all the SFR deployed are involved in the transmutation. All the other assumptions are the same as for Scenario 1. SFR are recycling MA coming both from LWR and SFR spent fuel. The minor actinides fraction is 10% and the matrix is UO_2. This fraction is the result of a compromise between:

- The fabrication and handling constraints of the blankets: effect of neutron sources, heat of fresh blankets (initial Cm fraction) and heat of irradiated blankets (burn-up, final Cm fraction).
- The necessity to recover the maximum of the minor actinides coming from PWR and SFR spent fuel reprocessing, so as to reduce the radiotoxic inventory of the high-level waste.

The radial blankets assumptions are displayed in Table 4.

Impact on the Pu inventory

Compared to Scenario 1, the Pu produced in the radial blankets allows deploying 3 GWe more at the end of the century (see Figure 4).

Table 4: Scenario 2 – radial blanket assumptions

Radial blankets	
Heavy nuclide mass per reactor	12.2 tonnes
Enrichment ($^{235}U/U$)	0.25%
Initial MA fraction	10%
Burn-up (MWd/t)	42 000
Reloading frequency	1
Cycle length (EFPD)	4 100

Figure 4: Scenarios 1 and 2 – SFR V0, two years cooling – total plutonium inventory

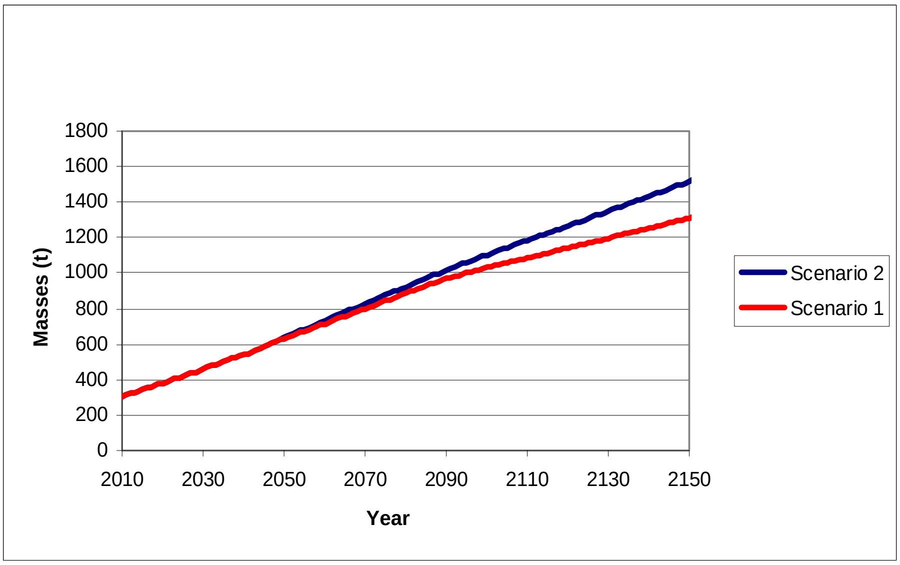
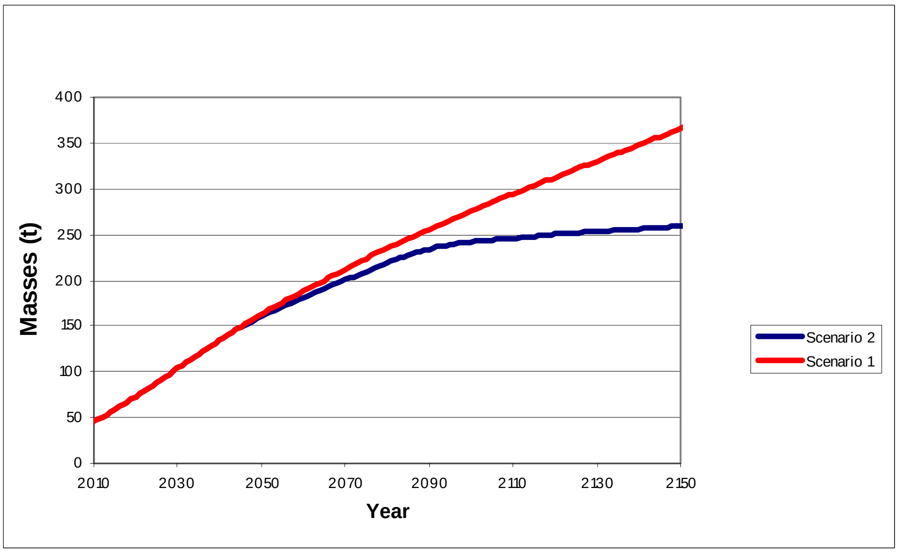


Figure 5: Scenarios 1 and 2 – SFR V0, two years cooling – total minor actinide inventory



Impact on MA inventory

The initial MA fraction in the radial blankets necessary to stabilise completely the minor actinides inventory in the fleet is linked to many assumptions or results as for example:

- The stabilisation of the plutonium inventory: an increase of the Pu inventory induces an increase of the americium inventory due to the decay of ^{241}Pu in the spent fuel storage.
- The cooling time of the spent fuel before reprocessing, for the same reasons.
- The recycling of the reprocessed uranium in the fast reactors, which induces an extra production of neptunium in the fast reactors.

For these reasons, an initial minor actinide fraction from 10 to 20% in the radial blankets is sufficient to stabilise the minor actinide inventory, with the condition to involve 100% of the SFR in the transmutation process. The optimisation of the transmutation scenarios is still ongoing.

Conclusions

The study conducted in 2008 has confirmed the previous studies. In the French electronuclear context, in the case of a scenario with a constant installed capacity, the plutonium margin obtained for the deployment of SFR is low, or negative if the minimum cooling time of SFR spent fuel is five years and if the SFR are deployed have a breeding gain close to zero. This margin decreases significantly when the ^{239}Pu equivalent rate in the SFR increases. For example, an increase of 10% of the equivalent ^{239}Pu mass in the core induces a decrease from 9 to 15 GWe for the possible installed capacity (no external source of plutonium is taken into account). In case of a lack of Pu, this lack appears in the second part of the SFR deployment after 2090, at the end of the transition period. The lack of Pu would be more important in the case of a scenario with an increase of the installed capacity.

It is possible to recover significant margins with the condition:

- to reduce the cooling time of the SFR spent fuel before reprocessing to three to four years in the period when the lack of Pu occurs, at the end of the century;
- to deploy SFR breeder (GRG = 0.2) with a small doubling time (approximately 50 years) early in the deployment of SFR (2040);
- to start the last SFR with an enriched U core instead of a MOX core;
- to reduce the burn-up of LWR fuel to increase the quantity and quality of the Pu produced;
- to deploy LWR instead of SFR in the last years of the deployment;
- to provide an external source of Pu;
- to stop the single recycling of Pu in the LWR.

The transmutation scenario of MA in the radial blankets allows deploying 3 GWe more, thanks to the Pu produced in the blankets. The MA inventory stabilisation can be reached with a MA fraction from 10 to 20%, depending from fuel cycle assumptions, with the condition to involve 100% of the SFR in the transmutation process. The optimisation of the transmutation scenarios is still ongoing.

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Transmutation of actinides in CANDU reactors

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Abstract

There is world-wide interest in reducing the burden on geological radioactive waste disposal sites. In most disposal scenarios, the decay heat loading of the surrounding rock limits the capacity of these sites. In the long term, this decay heat is generated primarily by the transuranic (TRU) actinides.

One approach to reducing this decay-heat burden is to reprocess spent reactor fuel and use thermal reactors to "burn" the actinides. The success of this approach is dependent upon the changes in isotopic composition of actinide-bearing fuels during irradiation in thermal reactor spectra. This work demonstrates the fuel cycle flexibility of the CANDU® reactor and its potential as an actinide burner to increase the capacity of geological radioactive waste disposal sites.

Many of the design features of the CANDU reactor make it uniquely adaptable to an actinide-burning role. The small, simple fuel bundle simplifies the fabrication and handling of active fuels. Online refuelling allows precise management of core reactivity. With the high neutron economy of the CANDU reactor, low fissile-loadings can be taken to high values of burn-up, providing high destruction of the TRU nuclides.

Several options for burning actinides in CANDU reactors have been investigated recently. One scenario mixes group-extracted transuranic nuclides with natural uranium to form a mixed-oxide (MOX) fuel, and then burns the material in a CANDU 6 reactor. Lattice cell calculations and full-core simulations have been done for actinide burning fuel in a CANDU 6 reactor. These simulations model spent nuclear fuel that was irradiated to 43 MWd/kg in a light water reactor and then decayed for 30 years. The full-core physics modelling code RFSP was used to perform the full-core calculations. This fuel was irradiated to a high target burn-up, resulting in 40% destruction of TRU. Various aspects of the reactor have been examined, including channel and bundle powers, and refuelling schemes.

A second option is to burn americium and curium, which are particularly problematic in nuclear waste disposal, in selected channels in the reactor. In these simulations, separated americium and curium are mixed with an inert matrix, and the outer channels of a CANDU 6 reactor are loaded with this material. The transmutation is driven by recycled uranium (RU) that comprises the rest of the reactor fuel.

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Introduction

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Many of the design features of the CANDU reactor make it uniquely adapted to undertake an actinide-burning role. The small, simple fuel bundle simplifies the fabrication and handling of active fuels. Online refuelling allows precise management of core reactivity. With the high neutron economy of the CANDU reactor, low fissile-loadings can be taken to high values of burn-up, providing high destruction of the transuranic nuclides.

This study expands upon work previously done [1-3] on mixed-oxide (MOX) fuels containing group-extracted transuranic elements. A second scenario is presented where americium and curium are transmuted in dedicated channels around the outside of the CANDU reactor.

Transmutation of actinides in MOX fuels

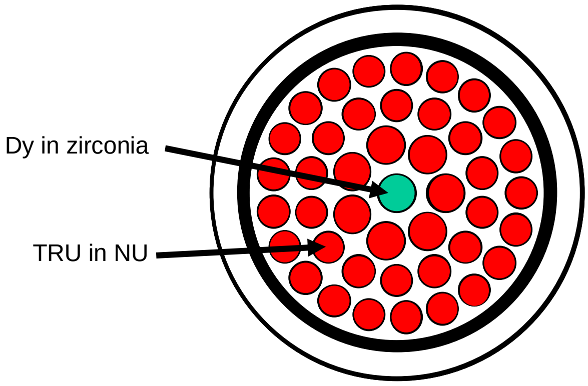
The input fuel simulated in this study was spent nuclear fuel from a light water reactor (LWR) that had been cooled for 30 years and then reprocessed to recover all the TRU [4]. The cooled TRU were then mixed with natural uranium to form a MOX fuel. This fuel is then transmuted in a CANDU 6 reactor. The input isotopic composition [1] of the fuel is given in Table 1.

Table 1: Input isotopic composition of the transuranic nuclides

Isotope	% by weight
^{237}Np	4.7
^{238}Pu	1.3
^{239}Pu	59.2
^{240}Pu	20.1
^{241}Pu	3.0
^{242}Pu	3.8
^{241}Am	9.9
^{243}Am	0.76
^{243}Cm	0.001
^{244}Cm	0.072
^{245}Cm	0.12
^{246}Cm	0.001

The fuel is 4.0% TRU by weight (heavy elements). The fuel bundle design used was the 43-element CANFLEX design, shown in Figure 1. The centre pin had a composition of dysprosium in a zirconia matrix in order to reduce coolant void reactivity.

Figure 1: Fuel bundle design for the MOX fuel



Lattice cell calculations were performed using WIMS-AECL [5] and full-core modelling used the Reactor Fuelling Simulation Program computer code (RFSP) [6]. The exit burn-up of the fuel was 43.4 MWd/kg(HE). This gave a total TRU transmutation of 42%. The percentage of each isotope transmuted is given in Table 2. Note that for this table a positive value is a destruction and a negative value is a creation. The input and exit compositions in grams per initial heavy metal metric tonne (IHMMT) are given in Table 3. There is an increase in curium mass, but it should be noted that most curium isotopes are short-lived and contribute to the decay heat on the same time scale as the fission products.

The evolution of the transuranic elements is shown in Figure 2. The high neutron economy of the CANDU reactor gives high values of destruction for the fissile nuclides. The fissile content of the TRU vector, initially 63%, drops to 30%. The support ratio (SR) is defined as the number of GWe output by LWR to 1 GWe output by CANDU reactors in a nuclear park to balance this recycling scheme. The support ratio for this fuel cycle is 11.2. The decay heat of the spent fuel is about 40% lower at 1 000 years after exit than LWR spent fuel that is not reprocessed and transmuted in a CANDU reactor. This could have significant impact on the capacity of a geological repository.

Full-core calculations of a MOX core have been completed to demonstrate the feasibility of fuelling a CANDU 6 core with this fuel. Several changes are needed with respect to the normal natural uranium (NU) fuel cycle. The fuelling scheme for NU generally involves replacing eight bundles in a fuel channel in each fuelling operation, referred to as an eight-bundle shift. Due to the higher initial reactivity of the MOX bundle, in order not to introduce too much localised reactivity into the core, the fuelling scheme is reduced to a combination of one- and two-bundle shifts. A schematic of the fuelling scheme is shown in Figure 3. One- and two-bundle shifts are applied to 264 and 116 channels, respectively. A detailed study on the impact on fuelling machine utilisation has not been completed, but the higher burn-up and therefore longer dwelling time of the fuel compensates for the smaller number of bundles shifted during refuelling. This results in a refuelling rate of about three bundles per day, compared to 15 bundles per day in the natural uranium CANDU 6 reactor.

A time-average model of the core was created to examine the maximum channel and bundle powers. In addition to this model, an instantaneous snapshot model was also examined. The instantaneous model, which generates random ages for the fuel channels, was used to analyse the power increase that occurs when refuelling the reactor, referred to as the refuelling ripple. Values for the maximum channel and bundle powers are given in Table 4.

Table 2: The amounts of the transuranic nuclides that are transmuted in this scenario

Nuclide	% transmuted
^{237}Np	51.0
Total Np	**48.1**
^{238}Pu	-206.6
^{239}Pu	78.6
^{240}Pu	2.4
^{241}Pu	-68.8
^{242}Pu	-128.2
Total Pu	**41.1**
^{241}Am	84.2
^{243}Am	-221.1
Total Am	**62.2**
^{243}Cm	-2 774.4
^{244}Cm	-1 676.5
^{245}Cm	-158.6
^{246}Cm	-1 706.0
Total Cm	**-2 374.4**
Total TRU	**41.9**

Table 3: Input and output amounts of actinides in grams per initial heavy metal metric tonne

Nuclide	MOX g/IHMMT	
	Charge	Discharge
^{234}U	51.5	32.0
^{235}U	6 783.6	1 235.1
^{236}U	0.0	868.9
^{238}U	947 332.2	927 272.0
Total U	**954 167.5**	**929 408.0**
^{237}Np	2 154.6	1 055.7
^{239}Np	0.0	59.8
Total Np	**2 156.6**	**1 118.3**
^{238}Pu	596.6	1 829.1
^{239}Pu	25 788.6	5 531.6
^{240}Pu	9 219.6	8 996.5
^{241}Pu	1 394.9	2 354.1
^{242}Pu	1 742.9	3 977.2
Total Pu	**38 742.6**	**22 688.5**
^{241}Am	4 543.3	716.6
^{242m}Am	0.0	7.2
^{243}Am	241.3	1 123.8
Total Am	**3 375.1**	**1 849.0**
^{242}Cm	0.00	355.2
^{243}Cm	0.5	13.2
^{244}Cm	33.1	587.2
^{245}Cm	5.5	14.2
^{246}Cm	0.5	8.3
^{247}Cm	0.0	0.1
Total Cm	**39.5**	**978.3**
Total TRU	**45 832.5**	**26 634.1**
Total HM	**1 000 000.0**	**956 042.1**

Figure 2: The isotopic evolution of TRU during transmutation in the CANDU reactor

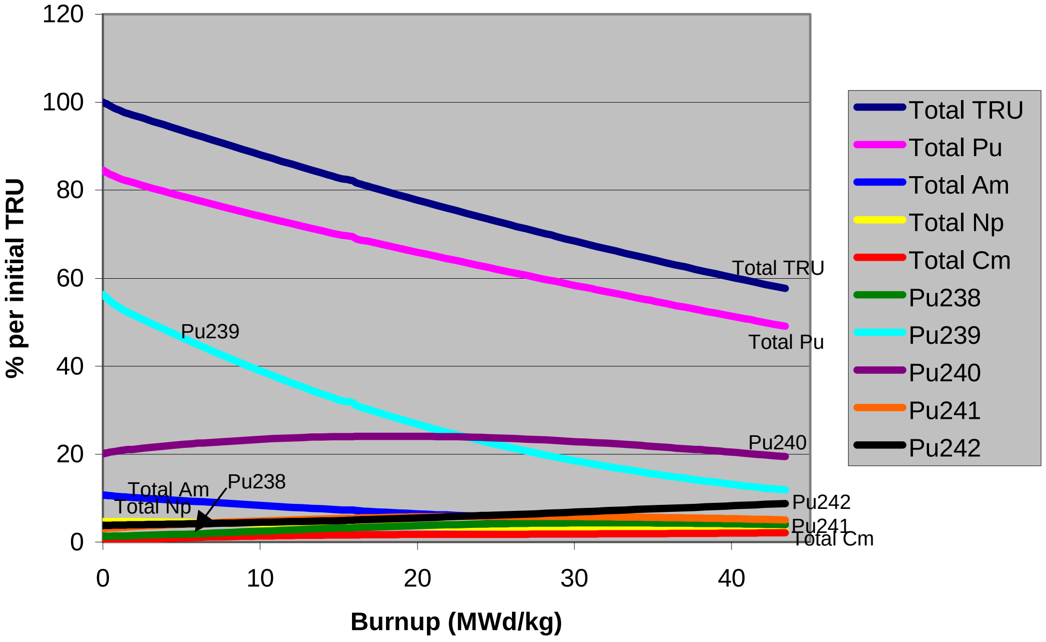
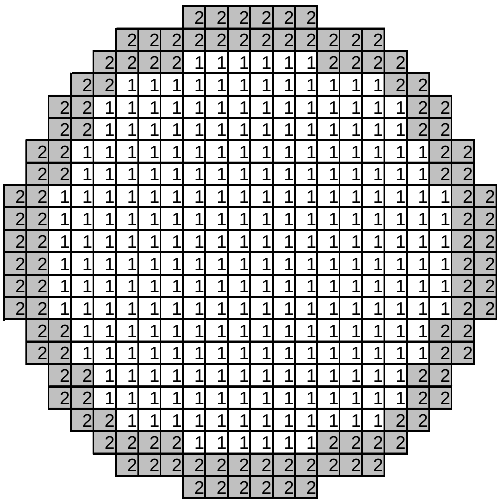


Figure 3: Schematic diagram showing the fuelling scheme for a CANDU reactor loaded with MOX fuel

The numbers 1 and 2 refer to channels with one- and two-bundle shifts, respectively



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								2	2	2	2	2	2								

Table 4: The maximum channel and bundle powers for the time-average and instantaneous models for the MOX fuel cycle

	Time-average model	Instantaneous model
Maximum channel power (kW)	6 300	7 200
Maximum bundle power (kW)	820	910

Americium and curium transmutation in an inert matrix

The fuel channel design of the CANDU reactor suggests the possibility of fuelling some channels with material to be transmuted and other channels with fuel to drive the transmutation process. In one such scenario, a CANDU 6 core was fuelled with 0.9% fissile reprocessed uranium (RU), while alternate channels around the outside of the reactor were fuelled with a mixture of americium and curium (AmCm) in an inert matrix fuel (IMF) (Figure 4). Several different amounts of AmCm in the fuel bundle were investigated: 14%, 19%, 26% and 35% by weight (897, 1 256, 1 794 and 2 691 g/bundle, respectively, for a CANFLEX fuel bundle design). Silicon carbide (SiC) has been used as the inert matrix material in this model. This work is equally applicable to a zirconia matrix, which would make an excellent material for direct disposal. The burn-up achieved for the RU is 12.2 MWd/kg(HE). This model makes use of leakage neutrons to transmute AmCm in low power channels. This is the maximum number of channels that could be used as targets and maintain criticality and symmetry in the core.

In this scenario, there are 30 channels of AmCm. The composition of the AmCm is taken from the Spent Fuel Composition database (SFCOMPO) maintained by the Nuclear Energy Agency. The data set used is Takahama-3 47.03 MWd/kg. The spent fuel was decayed for 30 years, and then the Am and Cm were separated out. The isotopic composition of the AmCm input to the CANDU reactor is given in Table 5. The cooling of the spent fuel leads to a significant increase in the amount of ^{241}Am, from the beta decay of ^{241}Pu.

Four different bundle designs were modelled for the AmCm transmutation targets. The bundle designs are CANFLEX, and three designs that are based on the 37-element design with pins removed: 21-element, 24-element and 30-element, see Figures 5 through 8.

Figure 4: Schematic diagram of the AmCm target model of a CANDU 6 core

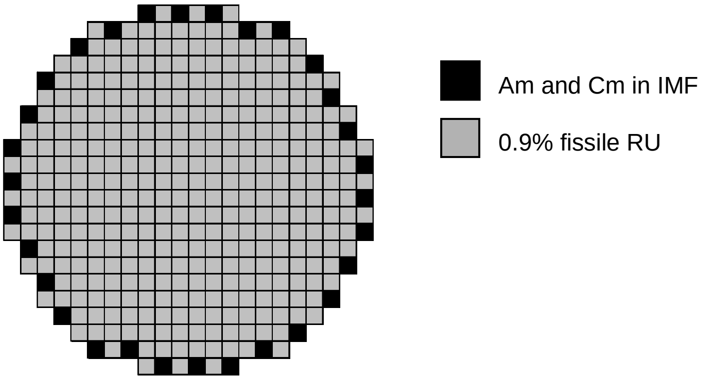


Table 5: Input composition of the americium and curium

Isotope	% by weight
^{241}Am	85.704
^{242m}Am	0.0666
^{243}Am	12.0214
^{242}Cm	0.0002
^{243}Cm	0.0268
^{244}Cm	1.7555
^{245}Cm	0.3783
^{246}Cm	0.0465
^{247}Cm	0.0007

Figure 5: Twenty-one element fuel bundle

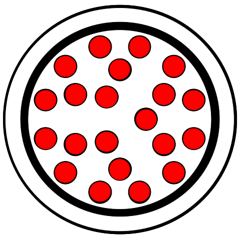


Figure 6: Twenty-four element fuel bundle

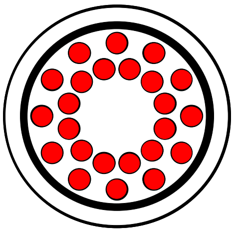


Figure 7: Thirty-element fuel bundle

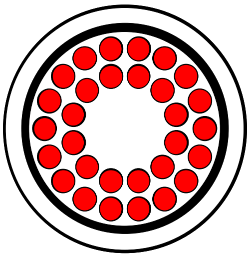
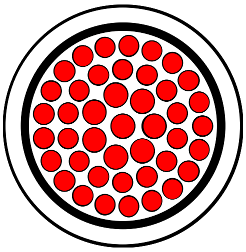
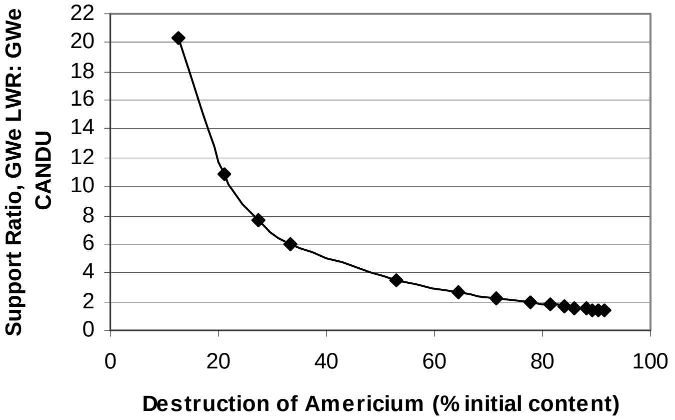


Figure 8: CANFLEX forty-three-element fuel bundle



A lighter fuel bundle, *i.e.* a bundle with fewer elements and therefore less mass, has a shorter residence time to achieve the same AmCm fractional destruction than a heavier bundle. However, with a lighter fuel bundle, less mass of AmCm can be fuelled in the reactor. The relationship between the support ratio[1,2] and the destruction rate of americium is shown in Figure 9. This plot is for the CANFLEX fuel bundle, but the relationship is the same for all of the bundle designs. The graph in Figure 10 shows the link between the percentage of AmCm that can be destroyed and the support ratio, and also how the heavier bundles require a longer residence time to achieve an equivalent destruction ratio. If a high support ratio is desired, then a lower destruction ratio is obtained, conversely if a high destruction of AmCm is desired then more GWe of CANDU reactors are required. A heavier bundle allows more AmCm to be input into the CANDU at one time, but it needs to burn longer to achieve the same AmCm destruction ratio. Therefore for a chosen destruction and support ratio the residence time can be chosen by selecting a lighter or heavier fuel bundle design.

Figure 9: The relationship between support ratio and destruction of Am for the CANFLEX bundle with 26% initial concentration of AmCm



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[1] The LWR reactor used for the spent fuel composition, Takahama-3, has a power of 2 652 MWth. A 33% efficiency was assumed (*e.g.* 875 MWe).

[2] A power of 725 MWe was assumed for the CANDU reactor.

Varying the initial amount of AmCm in the bundle produces the same relationship as shown in Figure 11. The amounts of AmCm that were modelled are: 14%, 19%, 26% and 35% by weight. This figure shows the effect of residence time on the destruction of AmCm and on the support ratio for CANFLEX fuel bundles with different input amounts of AmCm. This provides another means to choose the fuel design, whereby the initial AmCm concentration of the bundle can be chosen for a particular fuel bundle design, support ratio, percent destruction and residence time.

Figure 10: The black lines show the per cent destruction of Am, vs. residence time for each of the fuel bundle designs. The grey lines correspond to the right-hand y-axis and show the relationships among support ratios (SR) for the four bundles.

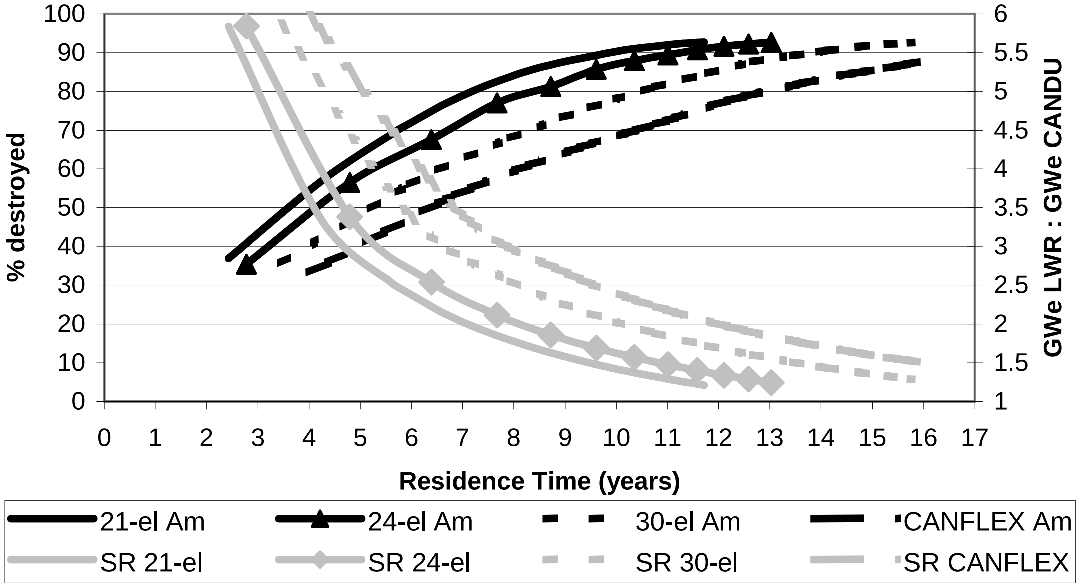
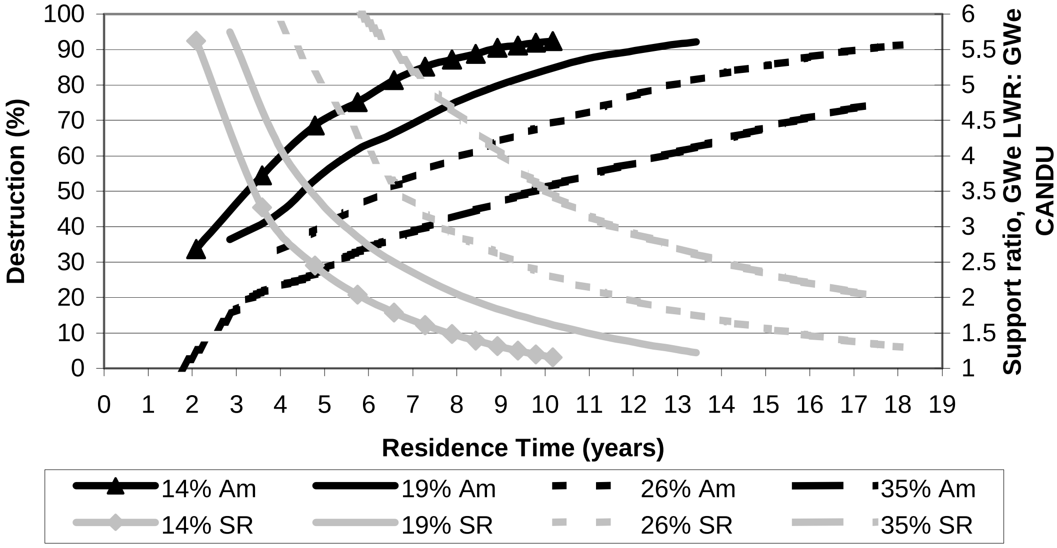
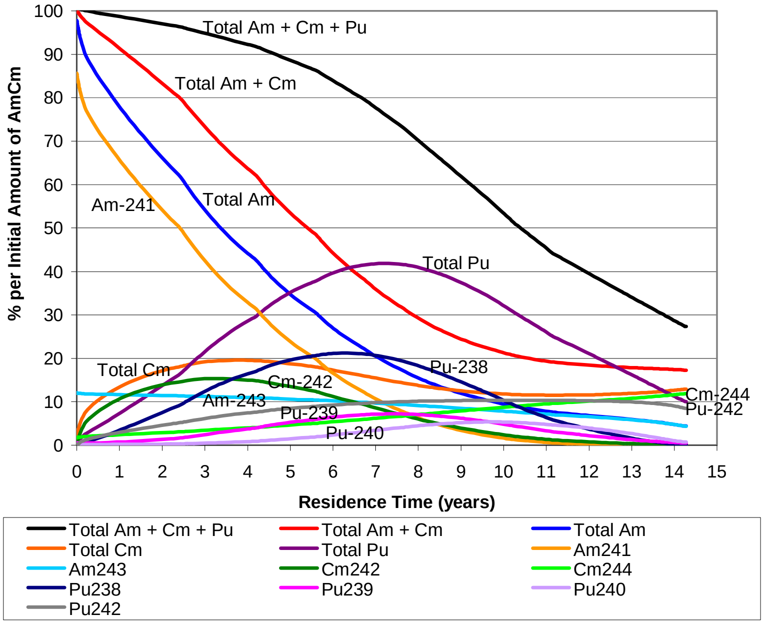


Figure 11: The black lines show the per cent destruction of Am, vs. residence time for each of the four initial concentrations of AmCm. The grey lines correspond to the right-hand y-axis and show the relationship among the support ratios (SR) for the four initial concentrations. All of these calculations were for the CANFLEX fuel bundle.



During the irradiation there is a growth of plutonium initially from the alpha decay of ^{242}Cm to ^{238}Pu, and from the electron capture decay of ^{242}Am to ^{242}Pu. The isotopic evolution of the AmCm is shown in Figure 12. The figure shows the results for the 21-element fuel bundle with an initial composition of 26% AmCm. Note that for any other case the trends are the same, but the scales on the axes will stretch or contract appropriately.

Figure 12: Isotopic evolution of the significant transuranic nuclides in the AmCm fuel for the 21-element fuel bundle with an initial composition of 26% AmCm by weight



Full core calculations have been performed for the AmCm target core using RFSP version 3.04.01. These models used a two-bundle shift for the AmCm and a combination of two- and eight-bundle shifts for the RU fuel. Table 6 gives the maximum channel and bundle powers for the time-average case and for the instantaneous model, which simulates a refuelling ripple for the CANFLEX bundle design with 26% AmCm initially in the bundle. These values are all within normal CANDU reactor operating conditions.

Table 6: The maximum channel and bundle powers for the time-average and instantaneous models for the AmCm target model

	Time-average model	Instantaneous model
Maximum channel power (kW)	6 660	7 080
Maximum bundle power (kW)	790	845

This transmutation of AmCm in targets in a CANDU reactor results in a decrease in decay heat at 1 000 years after exit from the reactor of 70% from spent fuel that was not transmuted in a CANDU reactor. This could have a substantial impact on the capacity of a long-term disposal facility.

The lattice cell calculations for the transmutation of actinides in MOX fuel were performed using WIMS-AECL. The lattice cell calculations for the AmCm target model used a developmental version of WIMS-AECL which has been modified to facilitate modelling depletion in targets with no substantial fissile content.

Both of the scenarios presented here, the full-core CANDU reactor loaded with MOX fuel, and the 30 peripheral channels loaded with AmCm in IMF, show promise as methods to transmute actinides. Both of these cases permit high throughput of actinides while requiring minimal changes to reactor design; that is, these fuel cycles can take place in existing CANDU reactors. The AmCm in IMF target model enables selected destruction of Am and Cm without breeding additional actinides. The use of an inert matrix may also have benefits for a disposal scenario as these actinides are already in a matrix and the fuel may not require further processing before disposal. The use of recycled uranium in the AmCm target model is important, as it is the main product from reprocessing. With these two scenarios CANDU reactors can utilise all actinide products from reprocessing: RU (AmCm model), minor actinides (AmCm and MOX models) and plutonium (MOX model).

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Further assessments of the attractiveness of materials in advanced nuclear fuel cycles from a safeguards perspective

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Abstract

This paper summarises the results of an extension to an earlier study [1] that examined the attractiveness of materials mixtures containing special nuclear materials (SNM) associated with the PUREX, UREX+ and COEX reprocessing schemes. This study focuses on the materials associated with the UREX, COEX, THOREX and PYROX reprocessing schemes. This study also examines what is required to render plutonium as "unattractive." Furthermore, combining the results of this study with those from the earlier study permits a comparison of the uranium- and thorium-based fuel cycles on the basis of the attractiveness of the SNM associated with each fuel cycle. Both studies were performed at the request of the United States Department of Energy (DOE), and are based on the calculation of "attractiveness levels" that has been couched in terms chosen for consistency with those normally used for nuclear materials in DOE nuclear facilities [2]. The methodology and key findings will be presented. Additionally, how these attractiveness levels relate to proliferation resistance (e.g. by increasing impediments to the diversion, theft, undeclared production of SNM for the purpose of acquiring a nuclear weapon), and how they could be used to help inform policy makers, will be discussed.

Introduction

The United States Department of Energy (DOE) requested an assessment of the attractiveness, from a safeguards perspective, of the special nuclear materials (SNM) associated with reprocessing. The metric used for evaluating the attractiveness of SNM is the following figure of merit (FOM):

$$FOM = 1 - \log_{10}(x)$$

where x is given by:

$$x = M\left[\frac{1}{800} + \frac{h}{4500}\right] + \left[\frac{D}{500}\right]^{\frac{1}{\log_{10}2}}$$

and M is the bare critical mass of the metal in kg, h is the heat content in W/kg, and D is the dose rate of $0.2 \cdot M$ evaluated at 1 m from the surface in rad/h.[1] The FOM is applicable to an adversary intending to build a stockpile of stable, threshold nuclear weapons using material in metal form without additional purifying other than what is assumed herein. An absolute scale by which to judge numeric values of the FOM is given in Table 1, which casts the FOM in terms of the utility of SNM for use in a nuclear weapon.

Table 1: Mapping of the FOM into utility for use in nuclear weapons

FOM for metals	Utility	Designation on plots
> 2	High	H
1-2	Moderate	M
0-1	Low	L
< 0	Off scale	O

In Ref. [1], the FOM was mapped into DOE Attractiveness Levels [2]. The DOE requires more stringent material control and accountability for higher Attractiveness Levels. Such a mapping is strictly not applicable, because the DOE Attractiveness Levels, given in Table 2, are determined by both quantity and quality of the SNM. Nevertheless, a meaningful correlation still exists between the FOM, Utility and Attractiveness Levels. From a safeguards perspective, SNM of high (H) utility (FOM > 2) is comparable in quality to material with an Attractiveness Level of B. Similarly, SNM of moderate (M), low (L), and off scale (O) utility are comparable to material with Attractiveness Levels of C, D and E, respectively. To avoid confusion for those dealing with SNM that must be safeguarded according to DOE regulations, the FOM henceforth will only be described in terms of utility for use in nuclear weapons. From a safeguards perspective, the products of an ideal reprocessing scheme would have utility L or O (*i.e.* FOM < 1).

Table 2: US DOE Attractiveness Levels

Attractiveness level	
B	Pure products
C	High-grade materials
D	Low-grade materials
E	All other materials

Background

The reprocessing schemes analysed to date and reported herein are: UREX, COEX, ^{238}Pu spiking, PYROX and THOREX. The dominant source of material for all of the reprocessing schemes analysed herein, except THOREX, is spent uranium oxide from pressurised water reactors (PWR) and boiling

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[1] In Ref. [1], the dose was expressed in rem/h rather than rad/h, as is done here. This distinction is only important for SNM with a significant (≥500 rad/h) neutron dose. None of the SNM in Ref. [1] meets this criterion. However, this study analyses Cm, which does meet this criterion.

water reactors (BWR). Such spent fuel is typically characterised by its burn-up, expressed in MWd/kg of initial heavy metal. The average burn-up of spent fuel in the USA historically has ranged from ~15 MWd/kg for BWR and from ~25 MWd/kg for PWR [3] to present-day values of 45-50 MWd/kg. The isotopic composition of spent fuel was generated with ORIGEN2.2 [4] for burn-ups ranging from 7.5 to 90 MWd/kg for the purposes of this analysis. The calculations of the required ^{235}U enrichment of the fuel charge and the spent fuel compositions are in good agreement with similar published results [5-11]. Also varied was the spent fuel age at the time of reprocessing relative to the time of discharge and the time after reprocessing.

A burn-up of 45 MW·d/kg was assumed for the calculation of the thorium spent fuel isotopics. The burn-up calculations were done using the TRITON module of SCALE 5.1 [12], and the decay after burn-up was done using ORIGEN-ARP [13]. The advantage of TRITON over ORIGEN is that non-standard fuels can be used in calculations. Three fuel mixes at charge that contained thorium were analysed: thorium with weapons-grade Pu (94% ^{239}Pu), reactor grade Pu (53% ^{239}Pu) and LEU (19.9% enriched ^{235}U). The percentages of the mixes were determined by achieving the same average k_{eff} as a LWR (4.08% enriched) at a burn-up of 45 MW·d/kg.

Results

A list of the UREX products is given in Table 3. The FOM results for the non-uranium bearing products listed in Table 3 are given in Figure 1. The products with the highest FOM (utility for use in nuclear weapons) are Pu and Pu+Np, which have the same FOM value. The FOM of both Pu and Pu+Np decreases significantly with increasing burn-up, because the concentrations of ^{239}Pu and ^{241}Pu (*i.e.* the isotopes with relatively high fission cross-sections) decrease and the concentration of ^{238}Pu, which is an intense heat source, increases with increasing burn-up. The age of the spent fuel at the time of reprocessing has only a minor effect on the FOM (*i.e.* the FOM increases slightly with increasing age). Heat is the primary proliferation barrier for Pu and Pu+Np.

The next highest FOM value belongs to TRU. The FOM of TRU decreases significantly with increasing burn-up, because the concentrations of ^{239}Pu and ^{241}Pu decrease with increasing burn-up. However, the FOM of TRU increases significantly with increasing spent fuel age, because ^{242}Cm and ^{244}Cm, which are intense heating sources, quickly decay away (their half lives are 163 days and 18 years, respectively).

In contrast, the FOM of Am increases with increasing burn-up, because of the build-up of ^{243}Am relative to ^{241}Am as the burn-up increases and because ^{243}Am produces less heat relative to ^{241}Am. The FOM of Am decreases with increasing age, because of the build-up of ^{241}Am relative to ^{243}Am with increasing age due to the beta decay of ^{241}Pu to ^{241}Am.

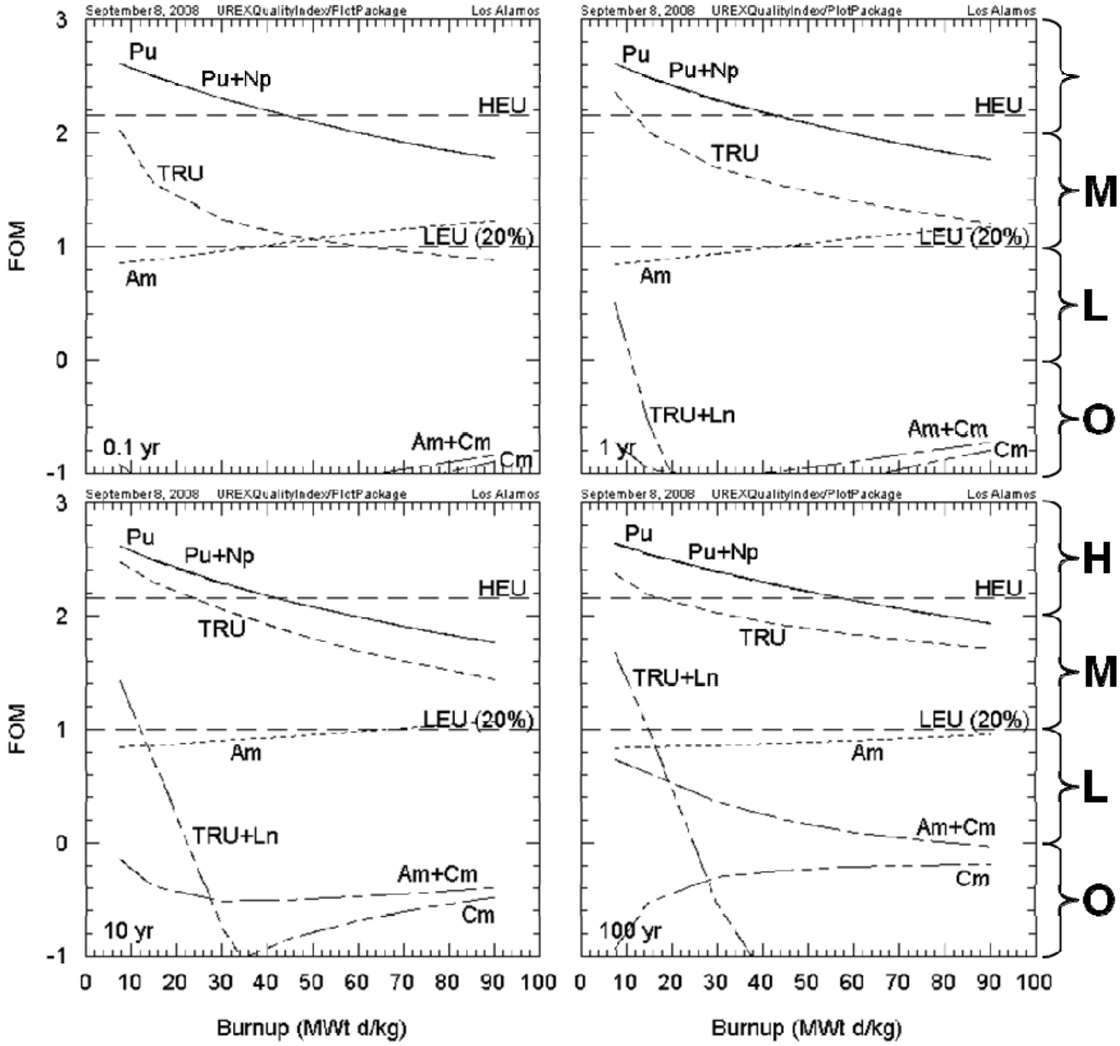
The FOM of Cm also increases with increasing burn-up, because of the build-up of ^{244}Cm relative to ^{242}Cm as the burn-up increases and because ^{244}Cm produces less heat relative to ^{242}Cm. As with TRU, the FOM of Cm increases significantly with increasing spent fuel age. Although Cm has a significant neutron dose, the FOM is dominated by its heating. Interestingly, the Am+Cm mixture has a low maximum utility, independent of burn-up and age.

Table 3: List of UREX products

Product	Process(es)
Pu+Np	UREX+2, UREX+3, UREX+4
Pu+Np+U	UREX+2a, UREX+3a, UREX+4a
TRU	UREX+1a
TRU+U	UREX+1b
TRU+Ln	UREX+1
Am	UREX+4, UREX+4a
Cm	UREX+4, UREX+4a
Am+Cm	UREX+3, UREX+3a
Am+Cm+Ln	UREX+2, UREX+2a

Figure 1: FOM of non-uranium bearing UREX products versus burn-up for various spent fuel ages at time of reprocessing

The letters H, M, L and O refer to the utility of the material for use in nuclear weapons (Table 1)



The retention of the lanthanides (Ln) with TRU or Am+Cm greatly reduces the FOM (the FOM of Am+Cm+Ln is so low that it does not appear in Figure 1), because the lanthanides provide an intense photon dose and, more importantly, an intense heat source. The FOM of TRU+Ln decreases with increasing burn-up, because of the relative build-up of lanthanides. The FOM of TRU+Ln increases with increasing age, because the lanthanides have half-lives that are of the order of 100 years.

The effect of diluting Pu and TRU with spent uranium is shown in Figure 2. The FOM is reduced with the addition of spent uranium. However, significant quantities of uranium are required to attain a low utility for use in nuclear weapons. For example, > 80% U is required for Pu, and ≥ 75% U is required for TRU, both obtained from 10-year-old spent fuel burned to 45 MWtd/kg. It should be noted that this study focused only on the utility of these uranium mixtures and did not consider any subsequent reprocessing by an adversary.

The effect that the period of time after reprocessing has on the FOM of Pu is shown in Figure 3. Increasing the post-reprocessing time decreases the FOM of Pu, because of the build-up of ^{241}Am from the beta decay of ^{241}Pu and the resulting increase in heating. As the age before reprocessing of Pu increases, there is less ^{238}Pu, which is the dominant heat source in Pu. Consequently, as the spent fuel age increases, the effect of the period of time after reprocessing decreases.

Figure 2: FOM of Pu (left) and TRU (right) versus spent uranium concentration for various burn-ups

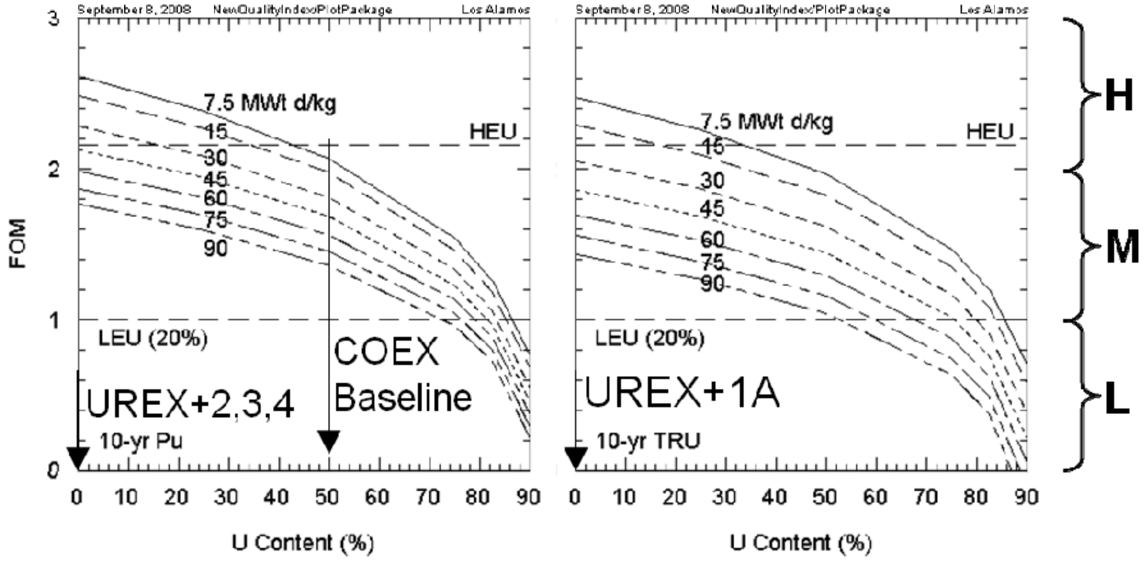
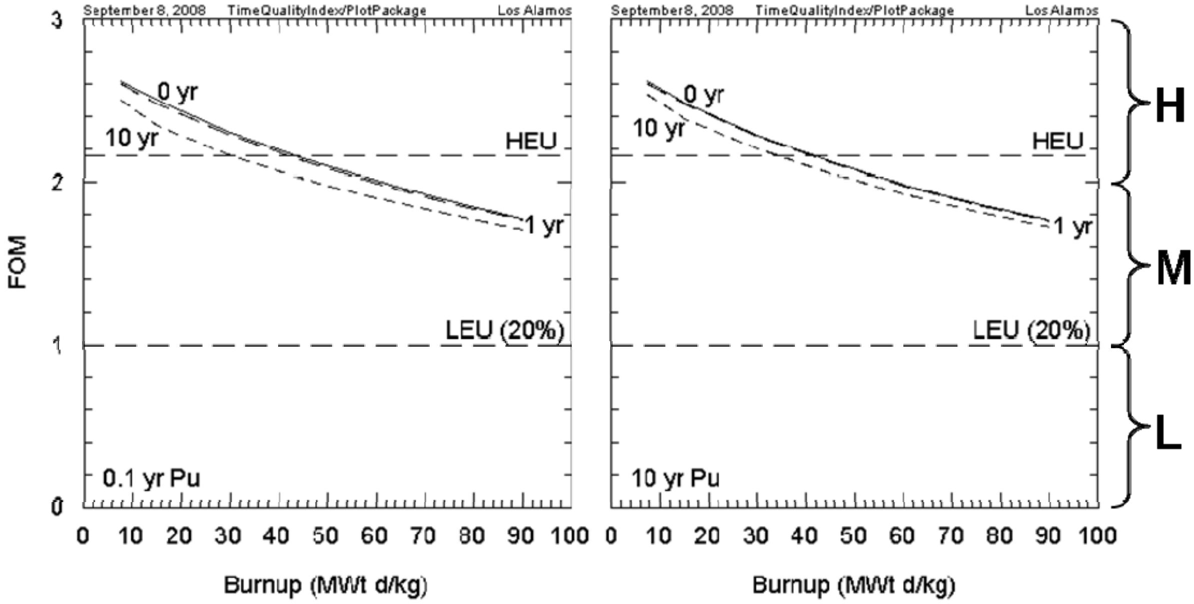


Figure 3: FOM of Pu versus burn-up for various spent fuel ages (lower left corner of each plot) and range of times after reprocessing (alongside of curves)



The effect that the period of time after reprocessing has on the FOM of TRU is shown in Figure 4. The behaviour of TRU is dominated by the concentration of ^{242}Cm and ^{244}Cm, which are intense heating sources that quickly (their half lives are 163 days and 18 years, respectively) dissipate. Then, for TRU taken from relatively fresh spent fuel, increasing the post-reprocessing time significantly increases the FOM. As the age of the spent fuel from which the TRU is taken increases, the change in the FOM with increasing post-reprocessing time decreases.

The effects of "denaturing" [14,15] the plutonium isotopic vector are shown in Figure 5. The plutonium in spent fuel has up to 8% ^{238}Pu, depending on the burn-up. Adding additional ^{238}Pu does reduce the utility of the plutonium for use in nuclear weapons. However, 80% ^{238}Pu is required to reduce the plutonium to low utility. A source for that much ^{238}Pu has not yet been identified.

Figure 4: FOM of TRU versus burn-up for various spent fuel ages (lower left corner of each plot) and range of times after reprocessing (alongside the curves)

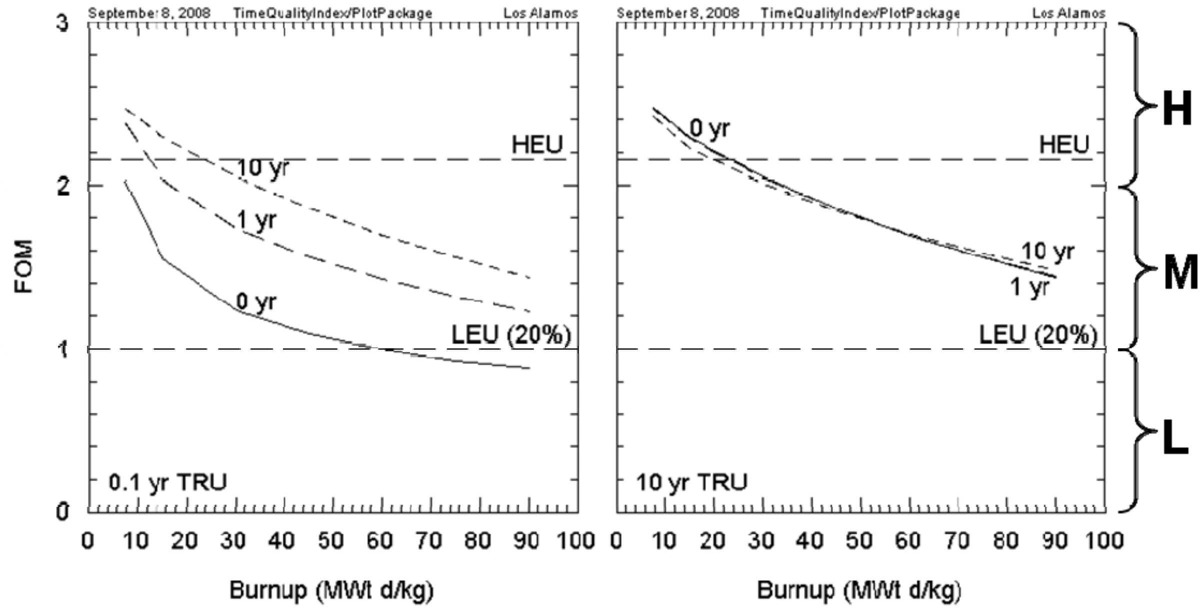
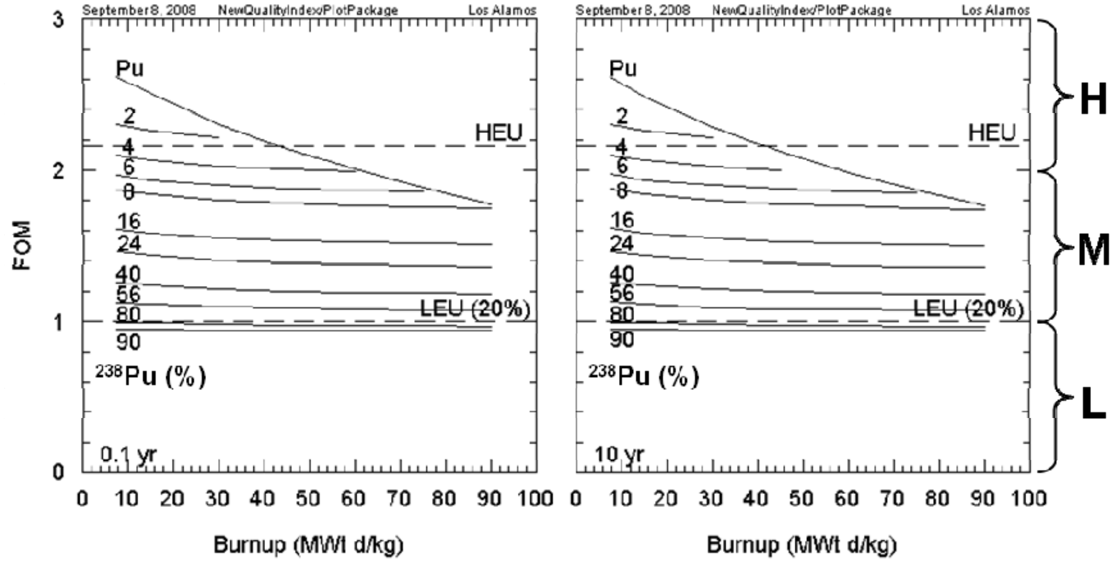
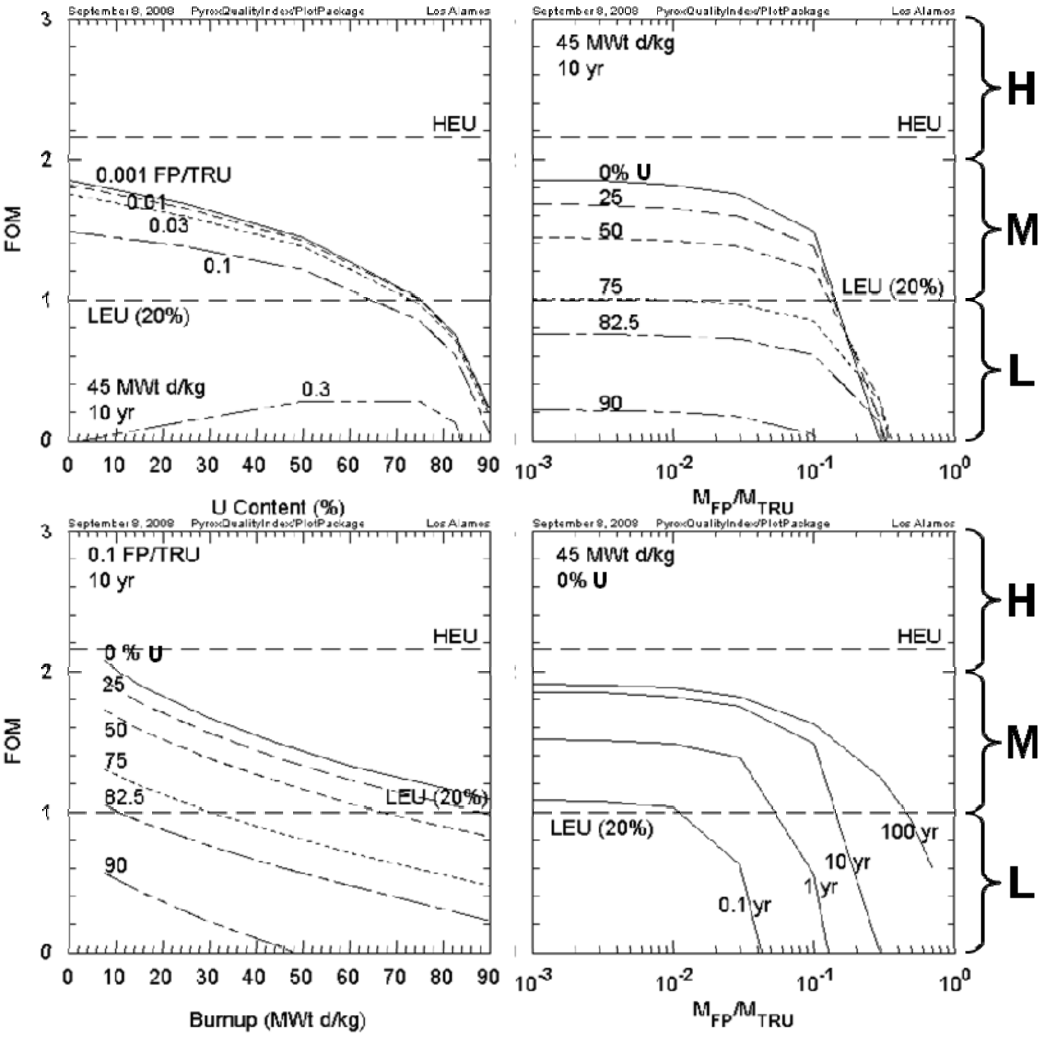


Figure 5: FOM of Pu versus burn-up for various concentrations (%) of added ^{238}Pu



The PYROX [16] products are U with trace (~100 ppm) amounts of TRU and a U+TRU mixture that is one-third U with small amounts (12 500 ppm) of the rare earth fission products. However, there are locations in the flow sheet where there is no U and a ~5% mixture of active metal and rare earth fission products. For this study, TRU with various concentrations of U and various concentrations of the active metal and rare earth fission products was examined. The active metal fission products will be removed in future work. The FOM for a range of mixtures of U, TRU and active metal and rare earth fission products is shown in Figure 6. The addition of increasing quantities of either U or active metal and rare earth fission products relative to TRU reduces the FOM. For a nominal reactor discharge that has been burned to 45 MWtd/kg and then cooled for 10 years before reprocessing, the TRU from that discharge would require ≥ 75% U for MFP/MTRU = 0 or require MFP/MTRU > 0.15 for 0% U to qualify as low utility, where MFP is the mass of the active metal and rare earth fission products and MTRU is the

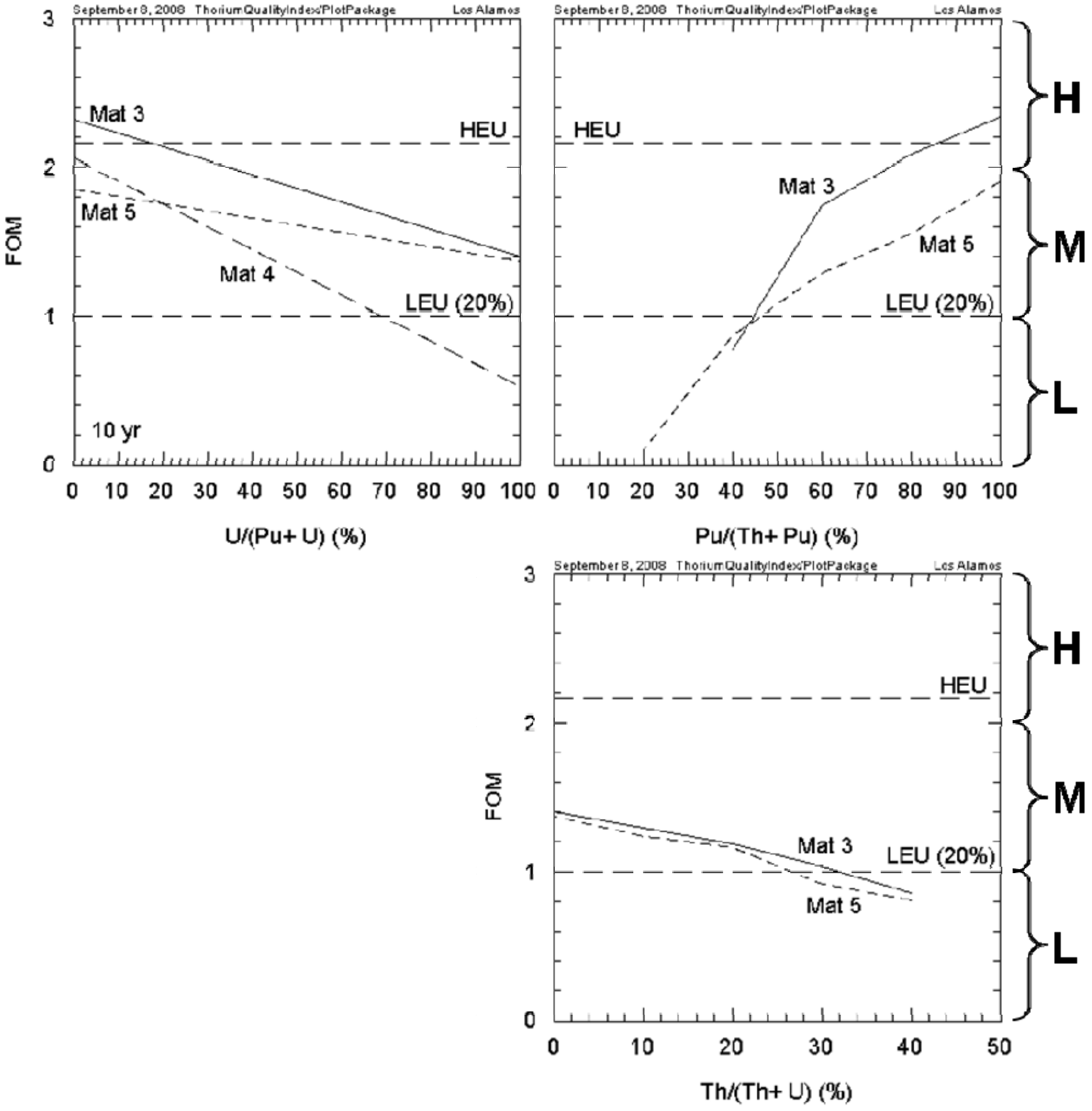
Figure 6: FOM of PYROX product versus uranium concentration (%), ratio of fission product (active metals and lanthanides) mass to TRU mass, and burn-up for various mass ratios, uranium concentrations and spent fuel ages



mass of the TRU. Similarly, various combinations of U and MFP/MTRU can be used to achieve a low utility. For example, 65% U and MFP/MTRU = 0.1 is also low utility. It should be noted that the value of MFP/MTRU in spent fuel ranges from ~1.0 for 15 MWtd/kg spent fuel to 3.0 for 90 MWtd/kg spent fuel. Several trends are also seen in Figure 6. Increasing the burn-up reduces the amount of U required for low utility. Increasing the age of the spent fuel at the time of reprocessing requires larger values of MFP/MTRU to achieve a low utility.

The thorium fuel cycle produces two isotopes that are of concern from a safeguards perspective: ^{233}U and ^{239}Pu. The ^{233}U is bred from thorium, and the ^{239}Pu is bred from any low-enriched, natural or depleted uranium that is introduced to dilute the ^{233}U that is bred from the thorium. For the three cases analysed herein, the ^{239}Pu is of greater concern from a safeguards perspective, as is shown in Figure 7. However, burning thorium fuel produces smaller quantities of ^{239}Pu than burning uranium fuel. Diluting the ^{239}Pu with spent thorium requires < 45% Pu (equivalently, > 55% Th) to achieve low utility. Diluting the ^{233}U with spent thorium requires > one-third Th to achieve low utility, depending on the initial quantity and quality of the Pu in the thorium charge.

Figure 7: FOM of THOREX product versus uranium, plutonium and thorium concentrations



Conclusions

The FOM of Pu+Np is the highest of the UREX products. The FOM of TRU, which is a UREX+1A product, is dependent upon spent fuel age and burn-up. Because the FOM of TRU increases significantly with spent fuel age, spent fuel should be reprocessed as soon as is practical. Furthermore, reprocessed TRU should be burned as soon as is practical, because the FOM of TRU increases significantly with increasing post-reprocessing time. For 10 yr, 45 MWtd/kg UREX+1A material (*i.e.* TRU), a U content > 75% is required to reduce the FOM to L.

The Pu+Np product has the same FOM as Pu product; coextracting Np with Pu does not reduce its utility for use in nuclear weapons. Conversely, extracting just Pu leaves Np in the waste stream. For 10 yr, 45 MWtd/kg COEX material (*i.e.* Pu), a U content ≥ 82% is required to reduce the FOM to L. The FOM of Pu (and Pu+Np) is not significantly affected by changing the post-irradiation time or by changing the post-reprocessing time.

The FOM of Pu denatured with ^{238}Pu concentration < 80% is still at least a utility of M. Based on the FOM formula used in this study, there is not enough ^{238}Pu (nor Np for breeding ^{238}Pu) to reduce the FOM to L.

The thorium fuel cycle produces two potentially attractive materials: ^{239}Pu and ^{233}U. The Pu is of greater concern from a safeguards perspective. The Pu product can be rendered unattractive by retaining > two-thirds Th fraction with it during/after reprocessing. The ^{233}U product can be rendered unattractive by adding natural or depleted U to the fuel before irradiation, but may exacerbate the ^{239}Pu problem in the product. Additionally, the ^{233}U product can be rendered unattractive by retaining > one-third fraction Th with it during/after reprocessing.

There is a safeguards and security benefit with respect to safeguards to diluting the reprocessing end products with Ln or reprocessed, natural or depleted U. However, there is no silver bullet to solve the safeguards and security issue. None of the proposed flow sheets examined to date justify reducing international safeguards or physical security protection levels. All of the reprocessing products evaluated to date must be rigorously safeguarded and provided the highest levels of physical protection.

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Session I

Impact of P&T on waste management and geological disposal

Chairs: V.P. Bhatnagar, M. Nutt

Concept of waste management and geological disposal incorporating partitioning and transmutation technology

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Abstract

To illustrate the benefit of partitioning and transmutation (P&T) technology, several types of the waste management and the geological disposal concepts incorporating P&T were discussed in terms of the repository size required to emplace the wastes. It was found that the transmutation of ^{241}*Am, which is a long-term heat source, is effective to prevent the inflation of the repository size expected in the case of plutonium utilisation. If we intend to reduce the repository size or enhance the capacity to a larger extent, for example by a factor of 4-100, in comparison with the conventional Japanese repository design for glass waste forms, the partitioning of Sr and Cs followed by their long-term (100-300 years) storage should be adopted together with the transmutation of MA. In such cases, to reduce the burden of the long-term storage of Sr and Cs, they are recommended to be contained in heat-resistant waste forms such as calcined wastes. The long-term storage without MA transmutation cannot achieve significant reduction of the repository size such as factor of 100 because of long-term heat from actinides.*

Introduction

Many studies have been made for the benefit of partitioning and transmutation (P&T) technology in terms of the reduction of long-term radiological toxicity, effect on the dose estimation for future inhabitants, reduction of the repository size, and so on. Among them, the recent tendency is to explore how much the P&T technology can mitigate the burden to find suitable sites for geological disposal by extending the capacity of the repository. From this viewpoint, the management of heat-generating nuclides in the high-level radioactive waste (HLW) is important.

After the reprocessing of the spent fuel, the influential heat-generating nuclides remaining in HLW are ^{90}Sr, ^{137}Cs and ^{241}Am, whose half-lives are 28.8 y, 30.1 y and 432 y, respectively. In Table 1, the heat generation of HLW are compared for various types of the nuclear reactors.

Table 1: Comparison of heat generation of high-level radioactive waste

Reactor type	Cooling time before reprocessing	Time after reprocessing	Heat generation normalised for 1TWeh	
			Fission products	Actinides
UO_2-LWR (43 GWd/t)	5 y	50 y	1 002 W	178 W
	20 y	50 y	703 W	355 W
	5 y	200 y	29 W	104 W
	20 y	200 y	21 W	258 W
MOX-LWR (43 GWd/t)	5 y	50 y	781 W	1 438 W
	20 y	50 y	548 W	2 078 W
	5 y	200 y	23 W	653 W
	20 y	200 y	17 W	1 364 W
MOX-FBR (79 GWd/t)	5 y	50 y	657 W	1 061 W
	20 y	50 y	462 W	1 611 W
	5 y	200 y	20 W	705 W
	20 y	200 y	14 W	1 192 W

The values in Table 1 were calculated for three kinds of spent fuels based on previous studies [1-3]. The first and the second types are UO_2 and MOX fuels from LWR, respectively. The burn-up was assumed as 43 GWd/tHM. The third type is for the MOX fuels from FBR, which was based on the 1 GWe FBR (79 GWd/tHM) presented in the benchmark exercise of different transmutation concepts conducted by the OECD/NEA [4]. The Pu isotopic composition of the fresh fuel was set by assuming to use Pu recovered from UO_2-LWR (50 GWd/tHM) after seven years' cooling. This type of spent fuel is, therefore, supposed to appear at the initial stage of the transient phase from LWR to FBR. The power generation efficiencies were assumed as 34.0% and 38.5% for LWR and FBR, respectively. Two cooling time periods, 5 and 20 years, between the discharge from the reactor core and the reprocessing were selected to understand the effects of the nuclear decay of the radioactive nuclides on the benefit of P&T technology. The decay heats at 50 and 200 years after the reprocessing are shown in Table 1. The values of the decay heats are normalised for unit generation of electricity (1 TWeh). The ORIGEN-2 code [5] was used for the burn-up and the decay calculation. ORILIBJ32 was used as the cross-section library for the burn-up calculation [6,7].

From Table 1, it is obvious that the heat generation of actinides is more influential than that of fission products except for the case of the UO_2-LWR at 50 years after the reprocessing. This fact suggests that if we remove the actinides from HLW and transmute them, the remaining fission products might be disposed of with a concentrated manner after a certain period of predisposal storage. On the contrary, if actinide elements remain in the HLW, it will take a long period of time to wait for enough decay of the heat-generating nuclides, particularly for the cases of the MOX fuels.

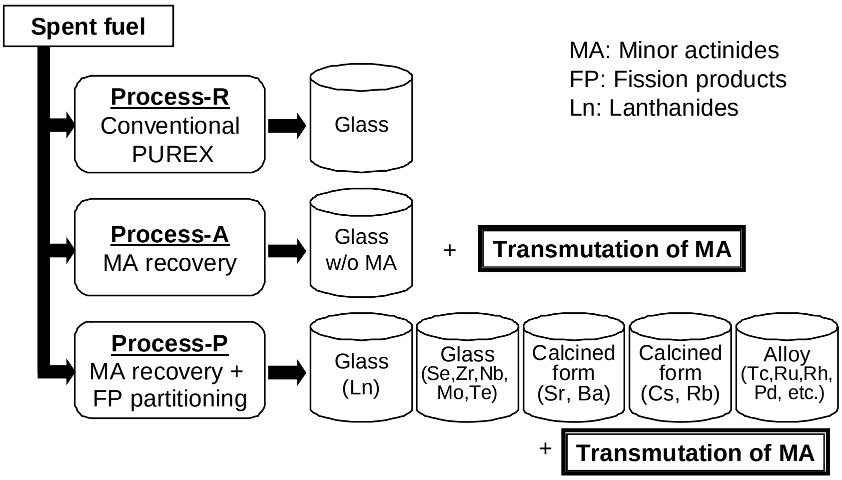
From the above-mentioned viewpoint, we recently investigated the relations between the time period of the predisposal storage and the repository area for the spent fuels from light water reactors [8]. It was revealed that the coupling of the MA transmutation, the FP partitioning and the long-term predisposal storage of Sr-Cs for about 300 years may provide us significant reduction of the burden of the waste management in terms of the repository size and the predisposal storage. The present study discusses the several types of the waste management and the geological disposal concepts incorporating P&T in terms of the repository size required to emplace the wastes from various reactor types shown in Table 1 and the time period of the predisposal storage.

Separation process

After the burn-up and the decay calculation mentioned in the previous section, the spent fuels were computationally separated by three optional processes: *ii)* conventional PUREX reprocessing (Process-R); *ii)* MA recycling without partitioning FP (Process-A); *iii)* full P&T for both MA and FP (Process-P). The schematic images of the wastes arising from these processes are shown in Figure 1.

In the conventional PUREX reprocessing (Process-R), the recovery efficiency of U and Pu was set at 99.5%. The conventional glass waste form was assumed as the HLW from this process. In the MA recycling without partitioning FP (Process-A), minor actinides such as Np, Am and Cm were assumed to be recovered and recycled in the FBR and/or the ADS. The recovery efficiency for these MAs was assumed as 99%, while that for U and Pu was 99.5%. A glass waste form containing FP and a small amount of MAs escaping from the recovery process was assumed as the HLW of this process. In the partitioning process of the full P&T case (Process-P), after recovering MA, FP was partitioned into five groups: *a)* lanthanides (Ln); *b)* precipitation at preprocessing containing Se, Zr, Nb, Mo, Te, etc.; *c)* Sr and Ba; *d)* Cs and Rb; *e)* Tc and platinum group metal (PGM). This separation scheme was based on the four-group partitioning process [9-11], and was modified to more detailed separation. The distribution ratios of major elements to the groups were listed in previous works [1,2]. The assumed waste forms were glass for groups *a)* and *b)*, calcined forms for groups *c)* and *d)*, and alloy for group *e)*, respectively.

Figure 1: Three options of separation processes and their waste forms



In this study, the HLW additionally arising from the transmutation of MA was not taken into account, because the amount of MA must be much smaller than the amount of FP in the spent fuel in general, and therefore the amount and the heat generation of the HLW from the MA transmutation process were considered comparatively small.

Estimation of amounts of waste forms

The numbers of waste forms for three kinds of fuel compositions, two kinds of cooling time periods and three kinds of separation schemes were estimated. The detailed conditions are described in Ref. [3].

As for the glass waste forms for Process-R and Process-A, the volume and the weight of one piece of glass waste form were assumed as 150 L and 400 kg, respectively. It was also assumed that one glass waste form can contain 60 kg (15 wt.%) of waste oxides and 12 kg (3 wt.%) of MoO_3. The maximum heat generation rate at the fabrication was restricted below 2.3 kW/piece. The amount of heat-generating nuclides in the glass waste form was also constrained so as to avoid the deterioration of the buffer material (bentonite) surrounding the waste forms from being heated up above 100°C. In the heat conduction analyses, the Japanese reference configuration for vertical emplacement of glass waste forms in crystalline rock was adopted; the pitch of the glass waste forms and the distance between the repository tunnels were fixed at 4.4 m and 10 m, respectively. The cooling period between the fabrication of the glass waste form and the emplacement to the repository was fixed at 50 years independently of the cooling periods before reprocessing. Forced cooling such as the ventilation after the emplacement of the waste forms was not taken into account.

As for the glass waste forms for Process-P, it was assumed that they can be high-density glass containing 140 kg (35 wt.%) of waste element oxides and 32 kg (8 wt.%) of MoO_3 at maximum. The heat generation rate of this glass waste form should be restricted under 2.3 kW/piece. The heat conduction calculation was not carried out because of its low heat generation.

As for Process-P, the calcined waste forms and the alloy wastes, which were not generated by Process-R and Process-A, should be managed. For the calcined forms, it was assumed that each form (14 L in volume) can contain 5.3 kg of Sr, Ba and other accompanying elements, or 4.5 kg of Cs, Rb and other accompanying elements. For the alloy wastes of Tc-PGM group, it was assumed that each form (7.5 L in volume, 60 kg in mass) can contain 2.4 kg (4 wt.%) of waste elements.

Table 2 shows the estimated numbers of the waste forms normalised by unit electricity generation (1 TWeh), The numbers of the glass waste forms for all the conventional PUREX reprocessing cases (Process-R) were determined by the temperature limit of the buffer material of the repository. When MA were recovered from HLW by the Process-A, the number of the glass waste forms was determined by the composition limit of waste oxides except for one case (UO_2-LWR after five years' cooling). When both MA and heat-generating FP were removed from the glass waste forms by Process-P, the numbers of the glass waste forms for "Ln" and "precipitation" were effectively reduced. Although the numbers of the calcined forms and the alloy wastes for Process-P seem large, their volumes are small. The areas needed to emplace these waste forms in the repository are discussed in the next section.

Table 2: Estimated numbers of waste forms per unit electricity production (piece/TWeh)

Reactor type	Cooling time before reprocessing	Process-R	Process-A
		HLW	HLW w/o MA
		Normal glass (150 L)	Normal glass (150 L)
UO_2-LWR	5 y	3.19*	2.65*
	20 y	3.12*	2.40
MOX-LWR	5 y	6.44*	2.36
	20 y	12.52*	2.36
MOX-FBR	5 y	6.02*	1.98
	20 y	10.25*	1.99

Reactor type	Cooling time before reprocessing	Process-P				
		Ln	Precipitation[†]	Sr, Ba	Cs, Rb	Tc-PGM
		High-density glass (150 L)	High-density glass (150 L)	Calcined form (14 L)	Calcined form (14 L)	Alloy waste (7.5 L)
UO_2-LWR	5 y	0.31	0.55	1.97	2.44	5.91
	20 y	0.31	0.55	2.11	2.17	5.91
MOX-LWR	5 y	0.29	0.50	1.87	2.65	8.32
	20 y	0.29	0.50	2.06	2.37	8.33
MOX-FBR	5 y	0.24	0.43	1.58	2.77	7.44
	20 y	0.24	0.43	1.73	2.55	7.45

* The number of waste forms was dominated by the temperature limit of the repository's buffer material after the disposal.

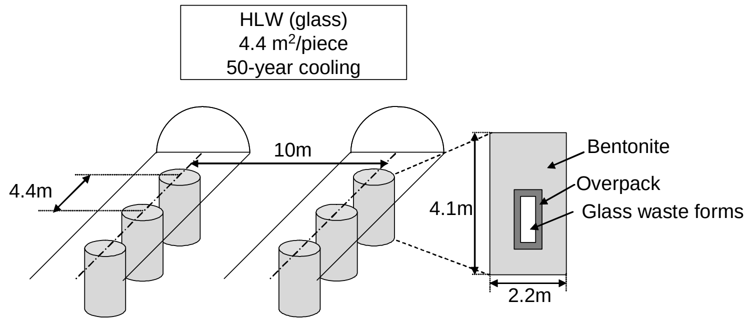
[†] Mainly containing Se, Zr, Nb, Mo, and Te.

Emplacement area for waste forms

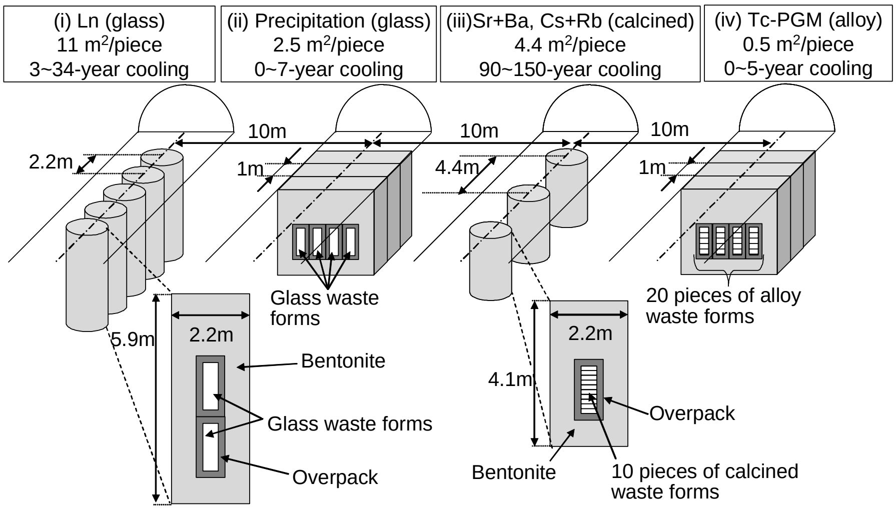
Because the number of the glass waste forms for Process-R was determined by the temperature limit of the buffer material, the repository area required for the emplacement of one piece of the waste form should be the same as the heat conduction calculation, namely, 4.4 m × 10 m = 44 m^2, as shown in Figure 2(a). The calculated areas of the deep underground repository required for HLW from 1 TWeh

Figure 2: Emplacement concept for various waste forms

a) Conventional emplacement concept for glass waste form



b) Examples of innovative emplace concepts for Process-P



of power generation are summarised in Table 3. The emplacement area of the glass waste forms for Process-R is not influenced by the cooling time. This is a result of compensation between the decay of heat-generating FP (^{90}Sr and ^{137}Cs) and the build-up of ^{241}Am. In the case of MOX-LWR for Process-R, the emplacement area is about twice as large as the case of UO_2-LWR for the cooling time of five years, while it becomes four times for the cooling time of 20 years. In the case of MOX-FBR, the required area is 10-20% smaller than that for MOX-LWR. This advantage of FBR is mainly caused by the good thermal efficiency of the FBR.

For Process-A, the emplacement area can be reduced in comparison with Process-R because of the removal of heat-generating MA from the waste forms. This reduction of the emplacement area by the MA recovery is remarkable in the cases of the MOX fuel for both LWR and FBR as shown in Table 3. It should be noted that the emplacement area per one piece of the glass waste form can be reduced further because the number of the waste forms was dominated by the composition limit instead of the temperature limit. In this study, however, the emplacement area was kept at 44 m²/piece and the possible reduction of the predisposal storage period was estimated. The reduction of the cooling time was about 10 years at maximum.

The high-density glass waste forms produced by Process-P can be compactly emplaced with short cooling time (3-34 y) as shown in Table 3. In this study, it was assumed that four times more compact emplacement of the waste forms was feasible from a viewpoint of the repository structure. An example of the emplacement concept is shown in the configuration-(i) of Figure 2(b). The high-density glass waste form for the precipitation can be emplaced much more compactly due to its low heat generation. In this study, about 18 times more compact emplacement than the reference concept was assumed as shown in configuration-(ii) of Figure 2(b). In spite of such compact emplacement, this waste form can be disposed of after a very short cooling time (0-7 y).

Table 3: Estimated emplacement areas for waste forms per unit electricity production (m^2/TWeh)

Reactor type	Cooling time before reprocessing	Process-R	Process-A
		HLW	HLW w/o MA
		44 m^2/piece*	44 m^2/piece*
		50 y†	40-50 y†
UO_2-LWR	5 y	140.4	116.6
	20 y	137.3	105.5
MOX-LWR	5 y	283.2	103.8
	20 y	550.8	103.9
MOX-FBR	5 y	264.9	87.3
	20 y	451.2	87.5

Reactor type	Cooling time before reprocessing	Process-P					
		Ln	Precipitation	Sr, Ba	Cs, Rb	Tc-PGM	Total
		11 m^2/piece*	2.5 m^2/piece*	4.4 m^2/piece*	4.4 m^2/piece*	0.5 m^2/piece*	
		3-34 y†	0-7 y†	90-150 y†	100-140 y†	0-7 y†	
UO_2-LWR	5 y	3.36	1.37	8.68	10.74	2.95	27.1
	20 y	3.36	1.37	9.29	9.54	2.96	26.9
MOX-LWR	5 y	3.14	1.26	8.24	11.66	4.16	28.5
	20 y	3.14	1.26	9.06	10.44	4.17	28.1
MOX-FBR	5 y	2.69	1.08	6.94	12.20	3.72	26.6
	20 y	2.69	1.08	7.63	11.21	3.72	26.3

* Emplacement area per one piece of waste form.

† Cooling period before disposal.

The disposal of the calcined forms containing Sr+Ba or Cs+Rb was a newly arising issue for Process-P. Since these waste forms are intense heat sources, we should wait for their cooling down. In this study, assuming that the ten pieces of the calcined forms (14 L/piece) are bundled to one package and disposed of as shown in configuration-(iii) of Figure 2(b), the time period to wait for the decay of the heat generation was roughly estimated. The criterion was set at 35 W/piece (*i.e.* 350 W/package), which corresponds to the criterion for the reference glass waste forms. The resultant cooling periods were 90-150 years for Sr+Ba, and 100-140 years for Cs+Rb. The emplacement area for one piece of the calcined waste form was then fixed at 4.4 m^2/piece. The area required for these new waste forms apparently dominates the total emplacement area for Process-P.

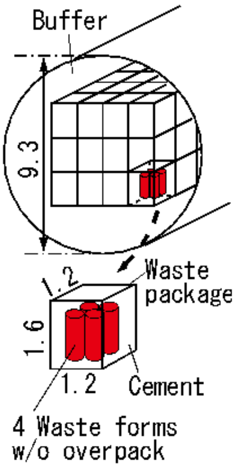
As for the alloy wastes arising from Process-P, it was assumed that twenty pieces of them were bundled and disposed of compactly as shown in configuration-(iv) of Figure 2(b). The emplacement area was then assumed at 0.5 m^2/piece. The cooling period was almost unnecessary.

Coupling with long-term predisposal storage

In Ref. [8], various emplacement configurations for the waste disposal were discussed by coupling P&T technology with long-term predisposal storage. In such configurations, the most compact one, shown in Figure 3, was based on the compressed waste forms of hulls and end pieces produced from LWR spent fuels. The emplacement area for this compact configuration was 0.95 m^2/piece for a waste form of 150 L. In the present study, the possibility to apply this compact emplacement configuration to various waste forms mentioned in the previous section is discussed. According to Ref. [8], the criteria of the heat generation to apply this compact emplacement is 4 W/piece. Table 4 shows the predisposal storage period required to wait for the reduction of the heat generation down to 4 W/piece, where it was assumed that 10 pieces of calcined waste forms (14 L) were bundled into one package and 20 of alloy waste forms (7.5 L) into one package.

All the glass waste forms containing MA (Process-R) r a storage period longer than 1 000 years. On the other hand, the waste forms without MA can be cooled down for very compact disposal. In the case of Process-A, 330-950 years are necessary, while about 300 years are commonly necessary for the calcined forms of Process-P.

Figure 3: Very compact configuration of waste disposal [8]



Distance between tunnels: 38m
0.95m²/piece

Table 4: Time period of predisposal storage required to wait for the reduction of the heat generation down to 4 W/piece

Fuel type	Cooling time before reprocessing	Process-R	Process-A
		HLW	HLW w/o MA
		Normal glass (150 L)	Normal glass (150 L)
UO_2-LWR	5 y	1 800 y	330 y
	20 y	2 600 y	330 y
MOX-LWR	5 y	6 000 y	600 y
	20 y	3 500 y	700 y
MOX-FBR	5 y	3 800 y	850 y
	20 y	3 200 y	950 y

Fuel type	Cooling time before reprocessing	Process-P				
		Ln	Precipitation	Sr, Ba	Cs, Rb	Tc-PGM
		High-density glass (150 L)	High-density glass (150 L)	10 pieces of calcined form (14 L × 10)	10 pieces of calcined form (14 L × 10)	20 pieces of alloy waste (7.5 L × 20)
UO_2-LWR	5 y	60 y	9 y	320 y	330 y	110 y
	20 y	45 y	0 y	310 y	320 y	100 y
MOX-LWR	5 y	75 y	9 y	320 y	330 y	70 y
	20 y	60 y	0 y	310 y	320 y	50 y
MOX-FBR	5 y	90y	10 y	320 y	320 y	70 y
	20 y	80 y	0 y	310 y	310 y	50 y

When the glass waste forms of Process-A are stored for long-term predisposal storage, Table 2 shows that about two pieces of glass waste forms (150 L) per 1 TWh should be stored. On the other hand, in the case of Process-P, it is necessary to store about four pieces of small calcined forms (14 L) per unit electricity production. The burden of long-term predisposal storage for Process-P, therefore, will be much lower than that of Process-A.

Table 5 summarises five typical concepts of waste management for UO_2-LWR. The conventional Process-R requires the emplacement area of 140 m² and the predisposal storage of 479 L of glass waste forms for 50 years. The adoption of Process-P reduces the emplacement area to 27 m², which is about one-fifth of Process-R, and 62 L of calcined waste forms should be stored for 130-150 years. When the long-term predisposal storage is coupled, the emplacement area is reduced to 1.5 m², which is

Table 5: Five typical concepts of waste management for HLW from UO_2-LWR (five years' cooling)

Normalised for 1 TWeh

Case	Waste	Waste form	Volume of waste forms	Predisposal storage	Emplacement area
Process-R	HLW	Normal glass	479 L	50 y	140 m^2
Process-A	HLW w/o MA	Normal glass	398 L	50 y	117 m^2
Process-P	Ln	High-density glass	46 L	18 y	3.4 m^2
	Precipitation	High-density glass	82 L	5 y	1.4 m^2
	Sr, Ba	Calcined form	28 L	130 y	8.7 m^2
	Cs, Rb	Calcined form	34 L	150 y	11 m^2
	Tc-PGM	Alloy waste	44 L	7 y	3.0 m^2
	Total		234 L	–	27 m^2
Process-A + long-term predisposal storage	HLW w/o MA	Normal glass	398 L	330 y	2.5 m^2
Process-P + long-term predisposal storage	Ln	High-density glass	46 L	60 y	0.3 m^2
	Precipitation	High-density glass	82 L	9 y	0.5 m^2
	Sr, Ba	Calcined form	28 L	320 y	0.2 m^2
	Cs, Rb	Calcined form	34 L	330 y	0.2 m^2
	Tc-PGM	Alloy waste	44 L	110 y	0.3 m^2
	Total		234 L	–	1.5 m^2

approximately two orders of reduction, and calcined waste forms should be stored for 320-330 years. If Process-A is adopted instead of Process-P to be coupled with the long-term predisposal storage, the significant reduction of the emplacement area can also be achieved, but the burden of the predisposal storage will be large. In order to judge which option is the most favourable, therefore, it is necessary to compare the cost of the partitioning process and that of the predisposal storage.

Conclusion

To discuss the benefit of P&T technology in terms of the repository size required to emplace the wastes, several types of waste management and geological disposal concepts incorporating P&T were proposed. It was found that the transmutation of ^{241}Am, which is a long-term heat source, is effective to prevent the inflation of the repository size expected in the case of plutonium utilisation. If we intend to reduce the repository size or enhance the capacity to a larger extent, for example by a factor of 4-100, in comparison with the conventional Japanese repository design for glass waste forms, the partitioning of Sr and Cs followed by their long-term (100-300 years) storage should be adopted together with the transmutation of MA. In such cases, to reduce the burden of the long-term storage of Sr and Cs, they are recommended to be contained in heat-resistant waste forms such as calcined wastes. The long-term storage without MA transmutation cannot achieve significant reduction of the repository size such as factor of 100 because of long-term heat from actinides.

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Summary of RED-IMPACT results on the impact of P&T on high-level waste management

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Abstract

RED-IMPACT is a research project funded by the EU within its 6th Framework Programme to explore the potential impact of partitioning and transmutation (P&T) and waste reduction technologies within advanced fuel cycles in the management of high-level waste.

The studies performed within RED-IMPACT show the potential value of P&T as a way to reduce the radiotoxic inventory over the long term and the heat source of the high-level wastes at the short and medium term. This last potentiality could allow significant enhancement of capacity of the final repositories. On the other hand, the studies show the small effect of these technologies on the dose levels to the public from the repository, under normal evolution conditions. Furthermore, the results of the studies indicate that the new intermediate-level waste of the advanced cycles need special attention, as they could represent an important fraction of the final radiotoxic inventory and volume to be disposed of in the final repository. This project also identified topics where further R&D is needed.

Introduction

Nuclear energy generates 30% of the electricity in the EU. Its characteristics, which will be further enhanced in the future generations of reactors, perfectly match the requirements of reduction of emissions, security of supply and competitivity, defined by the EC in the SET plan as the priorities for the future energy supply in the EU. Still, different countries of EU27 have very different attitudes towards the future use of nuclear energy for electricity generation or other uses.

Independent of the political decision of continuation or phase-out of nuclear energy, all countries using nuclear energy to generate electricity are facing the question of the final management of its spent nuclear fuel and other high-level radioactive wastes (HLW). Disposal in stable deep geological formations has been proven to be a technically viable solution to handle the already available and future spent fuel or HLW. However, deep geological disposal has not been fully implemented in any country, although Finland and the USA have approved its construction and identified, at least, one accepted site for the facility. Other countries like Sweden and France are also close to identifying potential sites, but most countries have not made such progress. Indeed, a special Eurobarometer of 2008 [1] shows that more than 70% of the EU27 population believes that "there is no safe way of getting rid of high-level radioactive waste" and only 43% out of the 79% that have an opinion, have understood that "deep underground disposal represents the most appropriate solution for long-term management of high-level radioactive waste." Finally, the study shows that the main fears are "the possible effects on the environment and health" (51%) and "the risk of radioactive leaks while the site is in operation" (30%).

In this framework, many countries have defined their present policy for waste management as the direct disposal of spent fuel, eventually after some interim near-surface storage for 40 to 150 years. However there are two arguments that are driving large interest on searching for viable variants and alternatives to direct geological disposal: the long-term sustainability of nuclear energy and the minimisation of the long-term legacy of hazards for future generations.

The spent fuel of the present light water reactors (LWR) contains very valuable materials, typically 95% of the uranium of the fresh fuel and plutonium with as much energy potential as 25% of the fissile part (^{235}U) of the fresh fuel. There are several reactor concepts, particularly fast reactors and ADS, that are able to use these components of the spent fuel to generate large amounts of electricity. These reactors will fission this new fuel to generate energy, transmuting it from long-lived actinides to fission fragments of much shorter half-life and with largely reduced long-term radiotoxicity. Some of these reactors are also able to use higher actinides (Np, Am and Cm) bringing further the utilisation of the energy potential and the minimisation of wastes from the spent fuel. A continuous recycling of actinides (U, Pu and minor actinides) that can be implemented combining reprocessing technologies with some advanced reactor concepts allows to multiply the amount of energy extracted per tonne of mined uranium by a factor between 30 and 100 [2] and to reduce the final high-level waste (HLW) to the reprocessing and fabrication losses (about a factor 1/100).

In order to reduce the amount of long-lived radioactive materials sent to final disposal, a process of separation and recycling, typical of many other industries, has been proposed. This methodology is generically described as partitioning and transmutation (P&T). The first step is to separate, or partition, the spent fuel into different components according to its final use or disposal requirements. Different options are considered for combinations but the basic components are: the irradiated uranium, the transuranium actinides, some selected short-lived (30 years) fission fragments (Cs and Sr), some selected long-lived fission fragments (I and Tc), the rest of the fission fragments, the activated structural materials and other intermediate-level wastes. The transuranium actinides contain most of the long-term radiotoxicity inventory and heat source, and along with the uranium most of the potential for additional energy generation.

After this partitioning some of these groups will be recycled in normal or advanced reactors (including subcritical ADS). In these reactors the actinides (U, Np, Pu, Am, Cm, …) undergo fission becoming fission fragments or are converted into other actinides, that is to say "transmuted". The spent fuel from this transmutation normally still contains significant amounts of actinides and it is necessary to repeat the partitioning and transmutation steps several times. In addition, the parasitic transmutation of few selected long-lived fission fragments could be done by neutron capture in these advanced reactors. The present studies indicate that very large reduction factors (typically 1/100) can be expected for the actinide inventories.

As a consequence of its potential benefits, large R&D initiatives on P&T had been launched world wide with large participation of the EU. The OMEGA project in Japan; the Accelerator Transmutation of Waste (ATW), Advanced Accelerator Applications (AAA), and Advanced Fuel Cycle Initiatives (AFCI), in the USA; a large partitioning and transmutation programme in the EU, and similar initiatives in Russia and South Korea had developed these new concepts. These projects have been efficiently co-ordinated in several NEA/OCDE working groups and IAEA projects. In parallel, the SNETP, Generation IV [3], GNEP and other similar initiatives have incorporated these concepts in their strategies.

Background on partitioning and transmutation

According to recent global energy scenario surveys [4-7,9], the increase in energy demand from emerging countries, the increase on cost of gas and oil and the need of limiting the emission of greenhouse effect gases will result in a future world nuclear installed capacity equal or higher to the present park. A standard light water power reactor of 1 GWe discharges about 23 tonnes of actinides each year; cumulatively 900 tonnes over a 40-year reactor lifetime. One tonne of spent fuel of average burn-up of 40 GWd/t contains about 10 kg of Pu and 1.5 kg of other transuranium elements, mainly neptunium, americium and curium, which are called the minor actinides (MA). These transuranium elements are not only long-lived radiotoxic substances but also major heat sources which affect the performance of the repository. It is predicted that without partitioning and transmutation of transuranium elements, “repository availability may be the major constraint to nuclear energy” [7]. The amount of spent nuclear fuel accumulated in Europe, is estimated at 37 000 tonnes for year 2000 with an additional 2 500 tonnes of spent fuel being produced every year [8].

A scientifically proven and technologically available solution for this spent fuel is its disposal in deep stable geological formations. Many countries including several from the EU have selected in the last decades the direct disposal of spent fuel as their final solution for these wastes, however none has yet built a geological repository and therefore no spent fuel has been directly disposed of thus far. The spent fuel generated is currently stored in the power plants (spent fuel pools or dry storage containers) or in centralised interim storage plants. Still, several EU countries like France, Belgium, the Netherlands, Germany and UK are or have been reprocessing part of their spent fuel.

In this context, a large R&D effort has been developed in the EU, and other countries, to evaluate and develop a complete set of partitioning and transmutation solutions. The EU effort has been co-ordinated around the different R&D framework programmes (FP) with a particular increase in the number of projects and resources in FP5 and FP6. A brief summary of the activities in FP4 and FP5 can be found in Ref. [11]. Complementary to the global evaluation of feasibility, performance, R&D needs and implications of the different options for the implementation of P&T in advanced fuel cycles had been performed in the framework of NEA/OCDE expert groups and the Working Party on Partitioning and Transmutation (WPPT) and the Working Party on Scientific Issues of Advanced Fuel Cycles (WPFC), and in IAEA projects. A comprehensive summary of most relevant projects, with detailed review of antecedents and a very complete list of references can be found in Refs. [8,12-14,17]. The first comprehensive report with a consensus of the possible performance and role of P&T as a waste management technology was Ref. [13]. This study however only evaluated the modification of inventories and expected consequences. The implications on the actual waste management and the potential benefits in the final geological disposal had first been studied in [14].

FP6 projects: RED-IMPACT

Those previous R&D efforts had been complemented in the FP6 by a number of projects including: technological aspects of the partitioning process in IP-EUROPART, technological aspects of the transmutation process in IP-EUROTRANS, the identification of R&D needs and the definition of a roadmap for this R&D in PATEROS, and the detailed evaluation of the impact of partitioning and transmutation in advanced fuel cycles for the reduction and final management of the HLW in the project RED-IMPACT [10].

The RED-IMPACT FP6 project includes a partnership of 23 organisations drawn from European nuclear industry (32%), waste agencies (18%) and research centres and universities (50%). The institutions were from 11 countries: Sweden (KTH), Germany (FZJ, FANP, GRS, IER, KKP), Belgium (BN, SCK•CEN), Czech Republic (NRI, RAWRA), EC (ITU-Karlsruhe), France (Areva ANP, CEA, COGEMA), the Netherlands (NRG), Romania (CITON), Slovakia (DECOM, VUJE), Spain (CIEMAT, EA, ENRESA) and the UK (NexiaSolutions, NIREX, UC).

The activities of RED-IMPACT have been structured into six work packages:

- review of waste management and transmutation strategies and selection of fuel cycles scenarios;
- feasibility of the industrial deployment of selected scenarios and their impact on waste management;
- assessment of waste streams, waste features, leach resistance, heat generation, reprocessing capability, etc. for selected fuel cycles;
- assessment of the benefits of P&T/C in advanced fuel cycles for waste management and geological disposal;
- economic, environmental and societal assessment of fuel cycle strategies;
- synthesis and dissemination of results.

With these work packages, RED-IMPACT covered most of the aspects needed to evaluate the impact of P&T in the waste management: scenario definition including reactors and other facilities, mass flows, waste streams for HLW and intermediate-level wastes (ILW), isotopic vectors, heat, radiotoxicity, radioactivity, and other parameters of the wastes in different points of the fuel cycle, waste forms and associated packages, repository concepts, evaluation of their relative size and capacity, and finally repository performance assessment and evaluation of doses.

Advanced fuel cycle scenarios

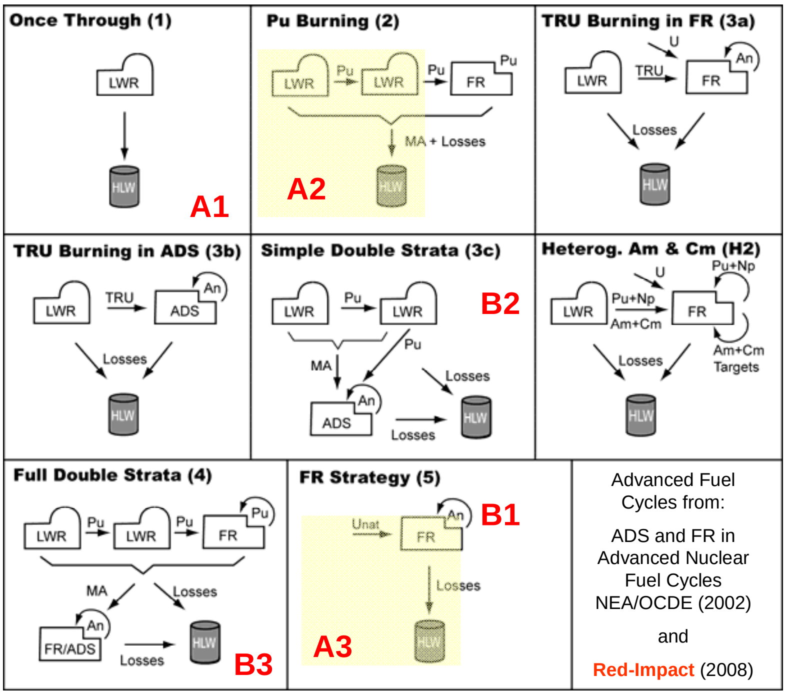
There are many possible new fuel cycle elements and combinations that had been proposed to implement the advanced technologies of P&T, each of these configurations is described in a fuel cycle scenario. In first approximation the critical parameters are the mass flows for the different actinides in the fuels or targets of each reactor included in the fuel cycle. Every single study has defined a new set of scenarios depending on the particular topics to be highlighted, however many conclusions are sufficiently generic so as to be valid for wide families of similar scenarios. A sufficiently wide set of scenarios was defined in the WPPT/NEA [13], and a subset was later adopted by the RED-IMPACT project, see Figure 1.

Indeed, within RED-IMPACT, after a comprehensive inventory of existing and foreseen nuclear fuel cycle facilities in Europe, it was possible to select a set of three so-called “industrial scenarios”, taking into account industrial feasibility of alternate strategies leading to increased actinide burning and reduced actinide generation: LWR with direct disposal (reference case A1), MOX fuel mono-recycling in LWR (A2), and plutonium recycling in sodium fast reactors (A3). In addition, three “innovative scenarios” were identified allowing multi-recycling of plutonium and MA and making use of advanced reprocessing technologies like DIAMEX-SANEX or COEXTM and PYRO processes: fast neutron Gen-IV system (B1), simplified double strata with LWR and ADS (B2), and double strata with fast reactors (FR), LWR and ADS (B3).

Between these scenarios there are, on one hand, those that transmute all the TRU, and on the other hand those that only recycle Pu (2), as in the simple closed cycle. Then we can identify scenarios that are based in a more or less complex single stratum (3a, 3b and 5), where the transmutation and the generation of electricity is done in the same reactors, and double strata scenarios (3c and 4) where the electricity generation is performed in reactors with clean fresh fuel (only U and Pu) and there are a small number of transmutation systems dedicated to the MA plus any remaining Pu. Another important element is the combination of systems with fast and thermal neutron spectrum and the selection for the TRU or MA transmutation of critical reactors or subcritical ADS. Finally the options of homogeneous transmutation of MA within the reactors fuels or their “heterogeneous” loading in specific transmutation targets (H2) had been analysed. Although not shown in this scheme the selection of the fuel nature and the reactor, recycling and fuel fabrication technologies also have an important influence on the scenario feasibility and performance. Indeed, the RED-IMPACT fuel cycles were selected to cover a wide range of representative waste streams, fuel cycle facilities and performances.

Figure 1: Advanced fuel cycle scenarios from WPPT/NEA [13] and RED-IMPACT [10] including LWR, FR and ADS

The figure indicates the flows of Pu, U, transuranic elements (Np, Pu, Am, Cm,…), TRU, minor actinides (Np, Am, Cm, …), MA or all the actinides (TRU+U), An



Transition scenarios

Apart from studying the scenarios described above in a state of equilibrium, a set of transition scenarios was also investigated in RED-IMPACT. The goal of doing so is first, to assess the feasibility of the selected scenarios and, second, to learn about the waste generation to provide basic information concerning strategic decisions with long-term effects. The three transition scenarios depart from a current nuclear power park and end in equilibrium situations as close as possible to a corresponding equilibrium scenario: A3, B1 and B2, see Figure 2.

A constant power production of 800 TWelh per year nuclear park, roughly corresponding to the present (2007) annual European nuclear power production, is assumed in these scenarios. At year zero (assumed 2010), the entire nuclear park consists of LWR, and the end of the transition period is fixed after 200 years. Year 2040 was selected as the year of industrial deployment of the new reactors, FR and ADS. The lifetime of all reactors is set to 60 years. The plant availabilities are postulated to 90% for all cores except the ADS, which is assumed to be of 70%.

Partitioning and transmutation expected performance

The potential benefits of P&T technologies depend first of all on the capacity of the various technologies and scenarios to reduce the inventories of critical components of the final high-level wastes as compared with the spent fuel of the open cycle. This performance was studied in detail in Ref. [13] for the eight different fuel cycles of the WPPT/NEA and by RED-IMPACT in its five scenarios, described in Figure 1. The results of the RED-IMPACT studies are shown in Figure 3.

RED-IMPACT estimates in 0.1% the actinides reprocessing losses, and consequently a reduction of more than 100 in the amounts of Pu and MA incorporated in the final HLW. The figure shows that different transmutation strategies could significantly reduce, *i.e.* one hundred fold, the radiotoxicity. This 1/100 factor is reached 400 years after fuel unloading for all the strategies that recycle and transmute plutonium and MA. If only Pu is recycled the factor becomes 1/10.

Figure 2: Evolution with time of the power share between different reactor types in the RED-IMPACT transition scenarios from A1 to A3/B1 (left) and to B2 (right)

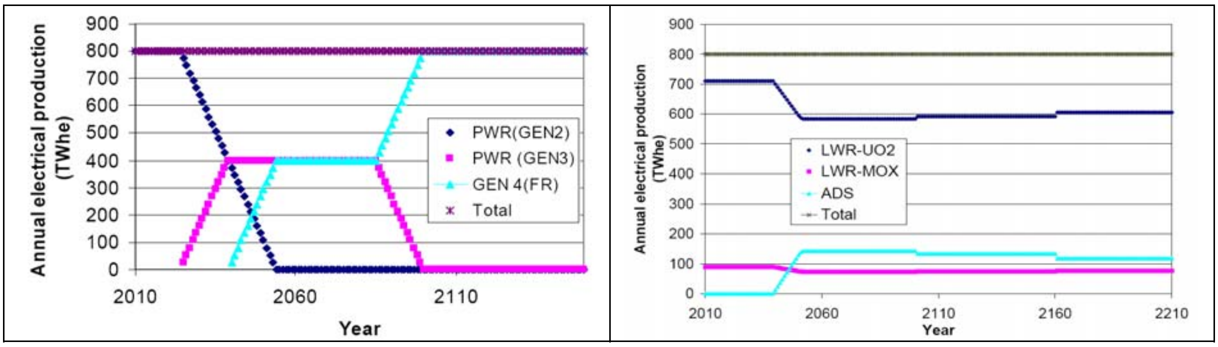
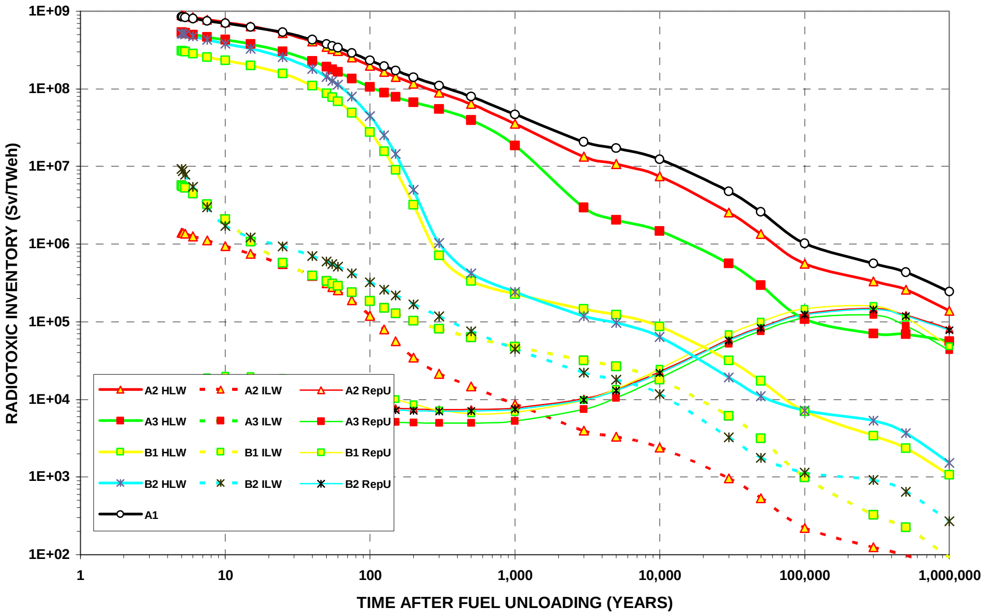


Figure 3: Radiotoxic inventories in the RED-IMPACT equilibrium scenarios from A1 to B2.The contributions from HLW, ILW and reprocessed U are shown separately.

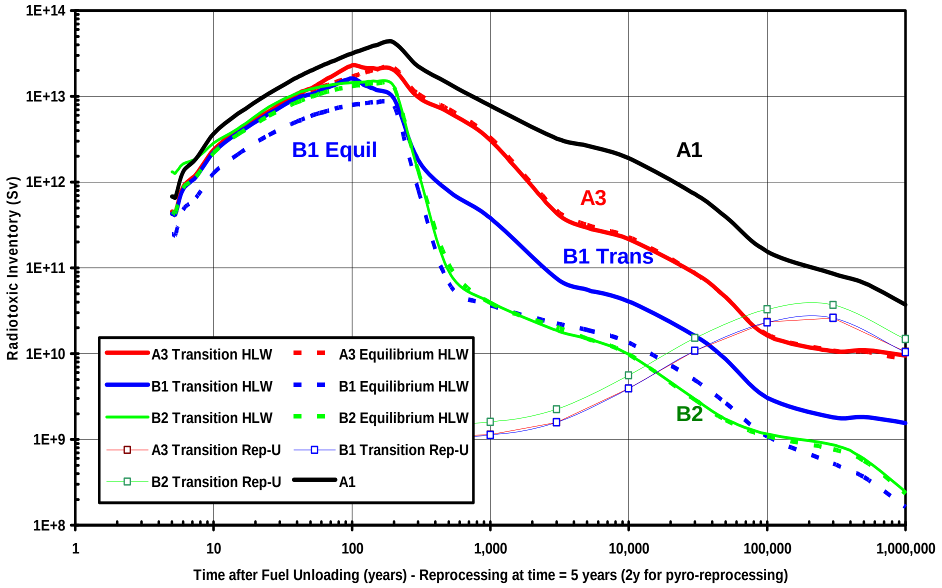


In addition the WPPT/NEA study concludes that as a consequence of the radiotoxicity reduction, while P&T will not replace the need for appropriate geological disposal of high-level waste, it improves the environmental friendliness of the nuclear energy option, and, in that respect, could contribute to a sustainable nuclear energy system. Very effective fuel cycle strategies, including both fast spectrum transmutation systems (FR and/or ADS) and multiple recycling with very low losses, would be required to achieve this objective. Fully closed fuel cycles may be achieved with a relatively limited increase in electricity cost of about 10-20%, compared with the LWR once-through fuel cycle.

The transition scenarios studies show that if the separation and transmutation technologies are applied when they are mature for their final performance the radiotoxicity reduction factors after 500 remain close to 1/100 (case B2 of Figure 4). However if the P&T process is applied when only Pu recycling is possible and MA are sent to the waste for some years (case B1 of Figure 4), the accumulated waste in this first period will limit the maximum performance by a significant factor (about 3 in the RED-IMPACT simulations).

These time-dependent studies have also shown the importance of a correct and sufficiently anticipated policy of P&T to take full benefit of the advantage of these advanced cycles. In particular, it is important to start the reprocessing early enough to have accumulated sufficient TRU, Pu or MA by

Figure 4: Radiotoxic inventories in the RED-IMPACT transition scenarios from A1 to A3, B1 and B2. The contributions from HLW, reprocessed U are shown separately.



the time it is needed in the transmutation plants, without requiring strong peaks in the utilisation of the reprocessing and fabrication plants. However, this will result in significant stocks of Pu and MA separated or in Pu/MA rich fuels fabricated in advance. The studies also show that reaching equilibrium composition is a very slow process and that significantly reducing the waste inventories in scenarios on reduction of nuclear power will require very long times, although regional co-operation [18,19] might reduce these periods to the lifetime of the transmutation plants.

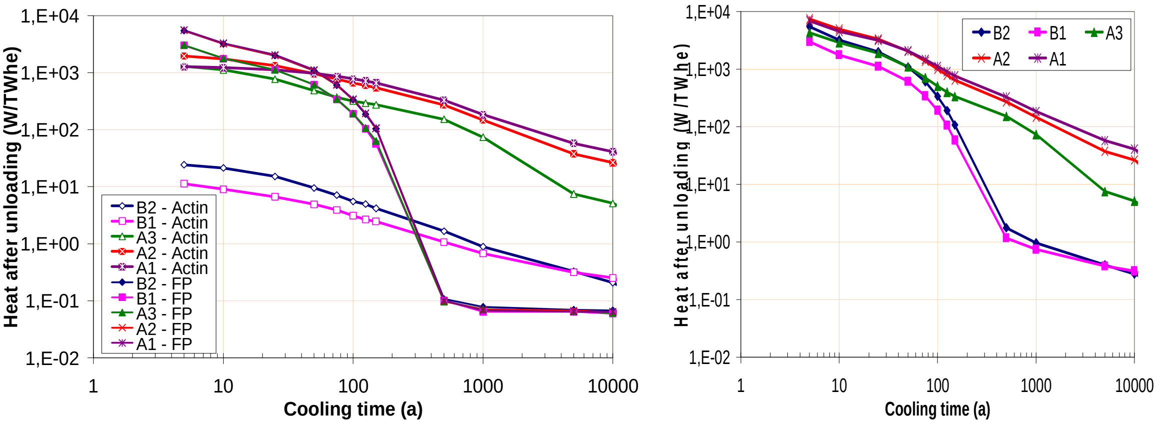
In addition, the transition scenarios have shown the relevance of waiting until the appropriate technology is available. For example, the use of standard reprocessing technologies that will send all MA to the HLW glasses will not be acceptable in scenarios of reduction of the nuclear energy installed power, where MA inventory reduction is one of the main objectives. If this were done, the inventory of MA immobilised in the glasses will limit the possible reduction on TRU mass to less than a factor 10 instead of the factor 100 reachable if the all the MA are transmuted before their storage. These considerations depend on the scenario, and this practice will be much more acceptable in the case of continuous or increased nuclear installed power.

Impact on the HLW thermal load

The reduction of the actinide content, in addition to the radiotoxicity and radioactivity, also reduces the thermal load from the waste. This heat load is a critical parameter for repositories on hard rock formations, strongly affecting its capacity. Figure 5 shows the heat load from the wastes of the different RED-IMPACT scenarios. In the LWR this heat load is dominated by MA after 100 years of cooling. As a consequence, in scenarios with recycling of MA the heat load is strongly reduced after 200 years. In addition, in these cases the heat load between 10 and 300 years is dominated by the 30-year fission fragments (^{90}Sr and ^{137}Cs).

Similar conclusions were obtained by RED-IMPACT and the WPFC/NEA study [14]: minor actinide management techniques, in fact, always reduce the total decay heat of the waste. While the heat reduction at normal disposal time (50 years) is modest (at most 70%), schemes with minor actinide management show a significant reduction potential at longer times. A ten-fold reduction could be achieved by prolonging the cooling time from 50 to 200 years. The same results can be obtained at the standard cooling times by separation and temporarily storage of Cs and Sr. The interim storage time needed varies within few decades depending on the waste loading. Additional cost of Cs/Sr separation and Cs/Sr interim storage should be taken into account. The WPFC/NEA study estimates at 5-10% the increase in total cost due to this interim storage. Removing and sequestering Cs and Sr in a separate

Figure 5: Heat load from wastes in the RED-IMPACT scenarios from A1 to B2.The contributions from actinides and fission fragments are shown separately on the left and the total in the right.



area of the repository or another facility would allow a further substantial increase in the drift loading of the repository, up to a factor of 43 in comparison with the direct disposal case for 99.9% removal of Pu, Am, Cs and Sr, according to Ref. [14].

Very similar conclusions have been reached in a number of studies concerning the separation and transmutation criteria to improve utilisation of a geologic repository applied to a repository similar to the one proposed at Yucca Mountain [15]. In these studies, the main conclusion is that the repository capacity, as defined by the different thermal limitations of the repository after the shutdown of forced cooling, can be multiplied by a factor of 5 if Pu and Am are recycled, by a factor 40 if Pu, Am, Sr and Cs are recycled and by a factor 91 if Pu, Am, Cm, Cs and Sr are recycled.

Impact on the deep geological disposal

The RED-IMPACT project has improved the realism of previous studies by including the evaluation of the intermediate-level wastes (ILW), and a more complete evaluation of the waste streams including structural materials and fuel impurities activation as well as the activation of all the specific components of the ADS (spallation target structural materials and coolant activation, spallation products activation, …). In addition RED-IMPACT took into account the implication of using presently available waste packages in the actual volume of the final wastes. Also the level of detail in the Cs and Sr handling scenarios and the performance assessment, including in this case the handling of the ILW, was improved. The RED-IMPACT [10] analysis allows reaching the following conclusions relevant for the impact of P&T on the repository capacity:

In scenarios where Pu or MA are largely included in the HLW, after 50 years of cooling the contributions from actinides dominate the thermal load of the HLW and consequently the evolution of HLW thermal power takes about 10^3 years to reduce by a factor 10 and about 10^4 years for a factor 100. On the other hand, for scenarios with full Pu and MA recycling, fission fragments dominate the thermal power for 300 years and so, the total thermal power for these scenarios is reduced by a factor 10 in only 100 years and by more than a factor 100 at year 300 after unloading.

These reductions show the possibility, for scenarios with full Pu and MA recycling, of large gains in the reduction of the thermal load to the repository and on its associated capacity by delaying the disposal time 100 or 200 years more. Similar reduction on the HLW thermal power can be gained at shorter times by separating the Sr and Cs from the HLW. In fact, when Pu and MA are recycled, the heat from the actinides plus 1% of these fission fragments, at the reference disposal time of 50 years, is only 2% of the total. This effect combined with the minimisation of fissile materials might help reduce the volume of the repository for granite and clay formations.

RED-IMPACT has shown that without specific Cs and Sr management, the HLW disposal gallery length can be reduced in factors that range from 1.5 to 6 for the granite and clay formations. If Sr is removed and Cs disposal is delayed by 50 additional years (disposal time was set as 50 years), then an additional factor 4 can be obtained in some of the geological formations, bringing the reduction in the disposal gallery length to a factor 13. Higher reduction factors are not excluded if the Cs disposal would be delayed longer.

On the other hand, RED-IMPACT has identified that the ILW might seriously compromise the advantages of P&T for the repository in some geological formations and for large reactor parks. In these conditions, sending these materials to geological disposals might require significant space and might generate additional dose that counterbalance the advantages of the HLW minimisation. The main contributors to the ILW maximum doses are the possible impurities in the cladding and structural materials, particularly ^{14}N that could be activated to ^{14}C. If this dose could become a significant hazard, the use of low activation steels and a stronger specification of the impurity content could possibly limit the problem. In any case, new repository concepts and, eventually, new legislation might be needed to properly handle the ILW, without handicapping the P&T advantages. Concepts of dedicated repositories for ILW at intermediate depth and with appropriated retention buffers are currently being developed by JNFL [16].

Impact on the performance assessment of deep geological disposal

RED-IMPACT results show that there is little to no advantage from P&T on the dose to the average member of the critical group from the normal evolution scenarios of the geological repository. This is expected because the main component for the dose in these scenarios is produced by fission fragments and activation products. On the other hand, P&T have a significant positive reduction on the dose to the reference group in the low probability human intrusion scenarios.

For all considered host formations, the impact of P&T on the maximum dose is limited, because the maximum dose is essentially due to long-lived fission products (Figures 6 and 7). One of the most important contributors to the total dose is ^{129}I. The amount of ^{129}I going into the repository as HLW very strongly depends on the fraction of spent fuel that is reprocessed.

For those long-lived fission products that pass to the HLW during reprocessing (^{79}Se, ^{126}Sn and ^{135}Cs), doses arising from the different scenarios are quite similar. For ^{129}I and ^{14}C and ^{36}Cl (activation products) the inventories in the HLW are much smaller than in the original fuel, because large fractions of those elements have been released as effluents. The decrease in doses due to ^{129}I, ^{14}C and ^{36}Cl in the scenarios with reprocessing is a consequence of the reduced inventory in the HLW. The transmutation of most actinides in fast reactors or accelerator-driven systems in case of advanced fuel cycle scenarios has little impact on the resulting doses, due to the low solubility of the actinides in reducing conditions and the strong sorption on minerals present in the buffer and host formation.

Figure 6: Estimated doses from the RED-IMPACT performance assessment of granite-H repository loaded with the spent fuel or HLW of the different scenarios from A1 to B2

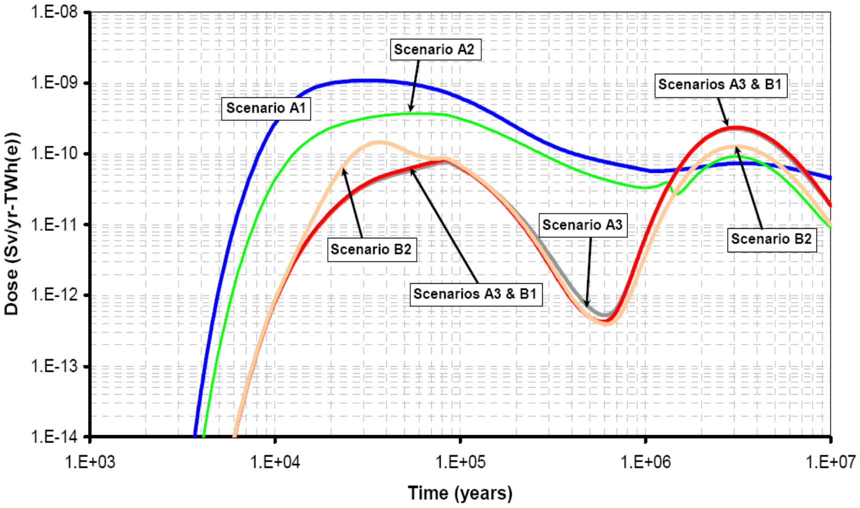
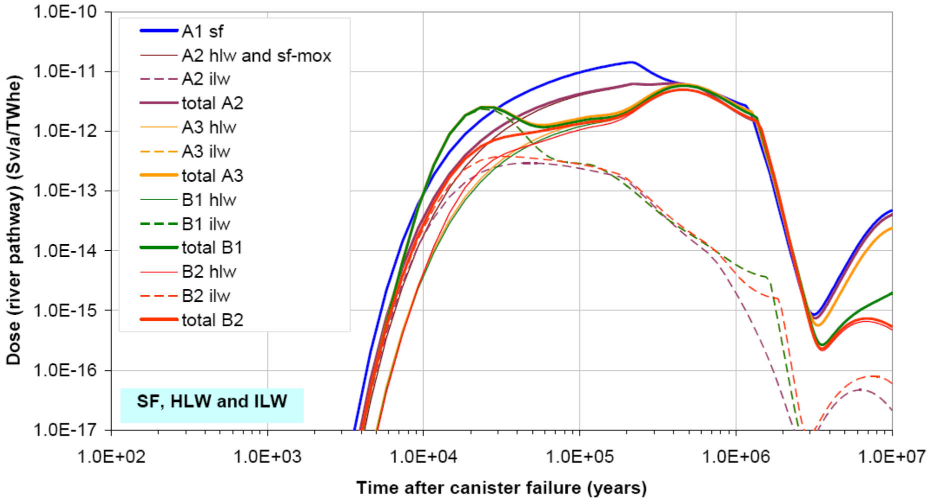


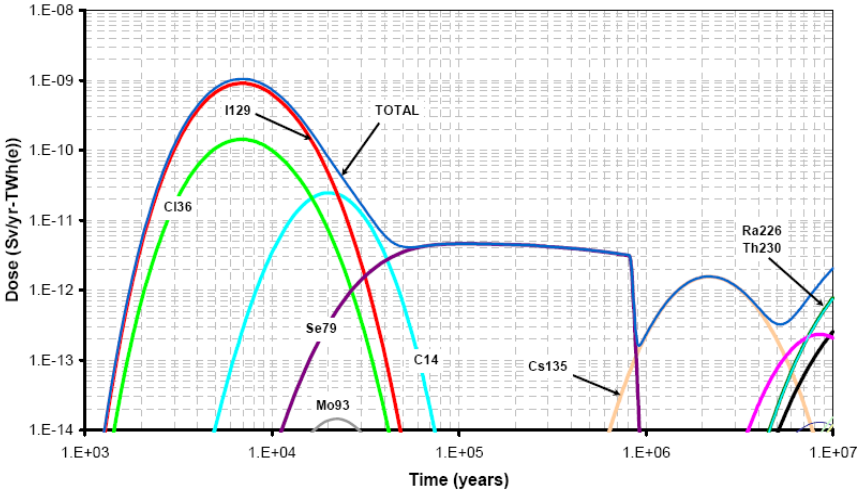
Figure 7: Estimated doses from the RED-IMPACT performance assessment of clay-B repository loaded with the spent fuel or HLW and ILW of the different scenarios from A1 to B2



When the waste disposal configuration is adapted to the thermal output of the disposed waste, the higher disposal density in case of advanced fuel cycles can result in a decrease of the release rate of solubility limited radionuclides (^{79}Se, ^{99}Tc and ^{126}Sn) in case of disposal.

Furthermore, the different scenarios are accompanied by different generation rates of long-lived ILW. The contribution to the biosphere doses by long-lived ILW is in some cases even higher than for HLW. The reason is that this type of waste is not assumed to be embedded in a stable matrix but more or less directly exposed to leaching after closure of the repository. In the evaluations made for disposal in granite, peak doses may exceed those of the HLW much earlier and may even dominate in the first ten thousand years (Figure 8). A lower contribution of long-lived ILW is expected for clay repositories (Figure 7). The high contribution of ^{14}C stemming from nitride fuels of fast systems and the activation of structural material as well as spallation products from ADS need to be included in an integral comparison. But, the contribution from the activation of structural materials can be minimised by suitable choices of structural materials in transmutation devices. Much more attention must be directed toward the generation of long-lived ILW and the management of long-lived fission and activation products including their conditioning and the development of repository concepts and appropriate engineered barriers.

Figure 8: Estimated doses from the RED-IMPACT performance assessment of granite-H repository loaded with the A2 scenario ILW



Human intrusion scenarios

An important argument in favour of geological disposal of the remaining HLW is that placing this waste deep underground is a robust method of reducing the potential for, and likelihood of, human intrusion, compared *e.g.* to the case of a surface storage. It cannot be guaranteed, however, that intrusion might not occur at some time in the future after administrative controls have been discontinued. As a complement to the safety and performance analyses, RED-IMPACT addressed the human intrusion, *e.g.* by a geotechnical worker into a deep repository for high-level waste and spent fuel types arising from five advanced fuel cycle scenarios considered within the project. The Cigar Lake uranium ore was used as a reference. This rock has a density of 2.8×10^3 kg/m^3 and contains 4.5×10^4 Bq/kg of ^{235}U and 1×10^6 Bq/kg ^{238}U. The comparison of the results indicates that the calculated dose for the high-level waste and spent fuel types, in the geotechnical worker human intrusion scenario, always exceeds the calculated dose from intrusion into the natural (Cigar Lake) uranium ore body for the assessment period of one million year duration. However, the radiotoxicity in the high-level waste or spent fuel as well as human intrusion doses after 500 years are drastically reduced by the transmutation of the actinides, exceeding in some scenarios a 1/100 reduction factor.

Conclusions

Partitioning and transmutation applies to the nuclear fuel cycle the general principle of most sustainable industries of classification (partitioning) and recycling (transmutation) of the components that are useful or dangerous for the population or the environment. With reasonable extrapolation of the performances of present technologies, P&T will allow largely reducing the long-term burden of the spent fuel and high-level waste, and can thus contribute to significantly improve its management. However, a deep geological repository to host the remaining high-level waste (HLW) and possibly some of the long-lived intermediate-level waste (ILW) will be needed whatever the procedure implemented to manage waste streams from different fuel cycle scenarios including P&T of long-lived transuranium actinides.

The main objectives of P&T are the transuranium actinides, neptunium, plutonium, americium and curium. P&T can provide a reduction larger than 100 on the mass of these transuranium elements sent to final disposal, reaching the same reduction factor on the radiotoxicity for the total of all the high-level wastes after a few hundred years. This large reduction of inventories provides a significant reduction of the consequences of low probability accidents, like human intrusion, and drastically reduces the potential proliferation interest of the repository. Inventory reduction also implies that the radiotoxicity reaches the level corresponding to the uranium mined for the fabrication of the fuel within 1 000 years, whereas the spent fuel in the open cycle will take several times 100 000 years to reach the same level.

The thermal load is the critical parameter determining the capacity of granite or clay deep geological repositories. The reduction of thermal loads from P&T can allow to increase the repository capacity or to reduce the disposal gallery length needed. If the HLW disposal is delayed by 100 years or Cs and Sr are separated by at least 100 years and P&T had been applied to all transuranium elements, the disposal capacity can increase by more than a factor 10, and in optimal conditions it is possible to reach a factor 50. Even if there is not such optimisation, the capacity can increase by factors from 2 to 6. These gallery length reduction factors can be used to reduce the cost of the repositories or to reduce the number of sites.

Special attention will be required for the specification of low impurity content in the structural materials and fuel, and to the optimisation of the intermediate-level waste management, as otherwise their increase in the advanced fuel cycles could seriously compromise the advantages of P&T for the repository in some geological formations and for large reactor parks.

P&T technologies will not significantly alter the dose to the average public person from the normal critical groups. Without P&T this parameter is designed to be more than one order of magnitude below the regulatory limits and the natural radiation background on the surface. This dose is mainly expected from the fission fragments and activation materials and so P&T will not provide significant reduction, except if iodine is transmuted. On the other hand, attention should be paid to avoid that the new ILW do not significantly increase this dose, particularly in the case of large reactor parks.

It must be noted that the benefits provided by P&T will not be obtained for free. Indeed if no optimisation, development and special protections are implemented, the risk of proliferation (reduced from the final repository) will increase in the different steps of the fuel cycle. Similarly the integral dose to workers in advanced fuel cycles could be larger. In addition, as already pointed out, there will be more secondary low- and intermediate-level wastes from the new steps of the advanced fuel cycles. Finally, depending on their implementation, advanced fuel cycles might require an increase in the transport of radioactive material, and the operation of interim storages of different radioactive streams.

P&T also requires the development of new technologies for many components of the nuclear fuel cycle: reactor technologies, accelerator technologies for ADS, fuel fabrication, advanced reprocessing technologies, coolant and material compatibilities. No real show stopper has been found in any of these fields, but the R&D effort needed is large and clear planning and intensive efforts are required to make these technologies industrially deployable by 2040-2050, when present estimations indicate they will be required. The PATEROS EU project has prepared a roadmap for this R&D to be developed in the EU, to identify, develop and demonstrate the options and technologies that could allow obtaining the benefits of P&T without unacceptable risks or costs in the advanced fuel cycles.

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Estimation of maximum permissible step losses in P&T processing

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Abstract

The main goal for any P&T process is to reduce a total potential hazard index of the deposited waste. Because the transmutation step normally reduces the amount of the irradiated element by a small amount, repeated P&T cycles are needed to reach the goal. In each cycle there is a small loss of material to waste streams. Traditionally, arbitrary requirements on the permissible losses have been used.

In order to calculate the requirements instead, we estimate the accumulated losses to wastes until nearly all material is either lost to wastes or destroyed by transmutation. By this procedure it is possible to set limits on the losses in each of the various steps during P&T recycling. Then it is also possible to compare these out-of-pile yield requirements for batch-wise and continuous P&T recycling.

Because the potential hazard index is different for different elements and their isotopes present during a P&T process the yield requirement is element specific.

A general, and perhaps surprising, result is that the yield requirements are much higher in a continuous P&T cycle than in a batch-wise recycling operation.

Introduction

In order to reduce the long-time potential hazard from finally deposited nuclear waste, or to reduce containment and space requirements for a final repository, an implementation of partitioning and transmutation (P&T) may reduce both amount and hazard from the treated long-lived elements originally present in the high-level waste. Irrespective of the separation and transmutation methods applied, a multiple recycling of the material involved in P&T is required in order to reach low final amounts of the treated elements. At the same time that material is separated and partially destroyed by transmutation, some of the material is also lost to various waste streams during each recycle.

Partitioning is normally able to separate, and concentrate, one or more chemical elements, but cannot normally separate isotopes. Hence the P&T process must be designed for chemical elements regardless of their isotopic composition. When the cycling losses to various waste streams during P&T are too large, almost all of the corresponding element will finally end up as waste. The P&T process has then in principle only generated a more dilute waste with almost the same total content of the long-lived isotopes as originally present before P&T operations began.

Because the majority of operating nuclear reactors are of the LWR type using fuel based on enriched uranium oxide or using smaller amounts of mixed-oxide fuel (UO_2+PuO_2) any implementation of a P&T strategy must adapt to this fact. It is then most feasible to envisage that P&T is implemented in order to destroy the minor actinides now present in the high-level waste stream from Purex reprocessing of the spent LWR fuel. Furthermore, it is also possible to envisage that neptunium is recovered separately in a modified Purex process. Hence, the elements of interest to remove from the HLLW stream are americium and curium. Uranium is usually not planned to be destroyed completely by P&T. Plutonium can be recycled as mixed-oxide fuel until its isotopic composition makes this operation unattractive. Remaining Pu should then join the minor actinides (Np, Am and Cm) in any P&T process used. For simplicity we will concentrate on Np, but the general calculations and arguments should be rather similar for plutonium, americium and curium.

Potential hazard-index of long-lived elements

The potential hazard of the long-lived elements in nuclear waste can be expressed as an index in several ways. We will use the hypothetical number of ALI:s in one kg of the waste dissolved in water as a base for our calculations. The conversion factor used for each radionuclide was taken from ICRP Publication 60 [1]. Now the hazard index calculated as a hypothetical number of ALI:s in the waste can be compared with the corresponding number of ALI:s from the uranium (including daughter nuclides) fissioned during the LWR operation generating the spent fuel which gave rise to the waste. Furthermore, for simplicity we assume that the biological availability of the deposited waste is about the same as for common uranium containing minerals at all times. Hence, a potential hazard index value lower than the uranium reference line represents a case with no long-time change in the danger to humans from long-lived nuclides in our environment.

The potential hazard index computed in this way will vary somewhat from reactor to reactor and from fuel type to fuel type. However, it can be used here as an illustrative base for our discussion of various flow sheets for P&T and their inter-comparison.

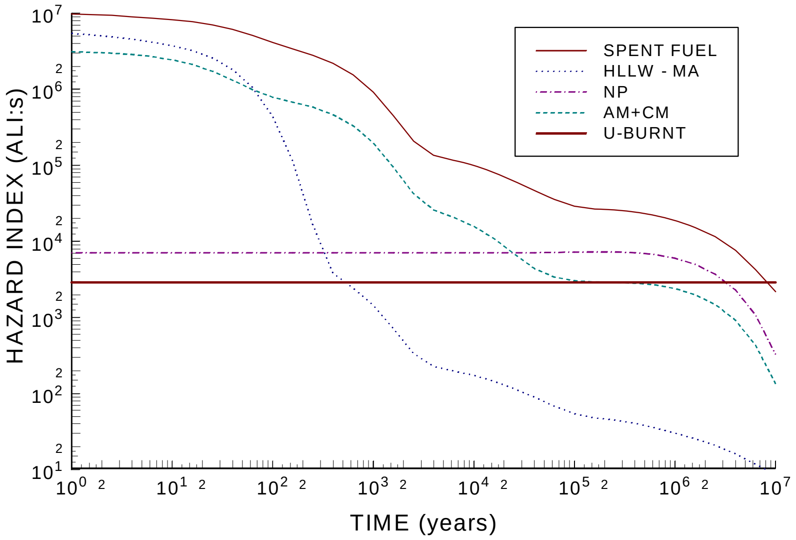
Figure 1 shows the calculated potential hazard in number of ALI:s as function of time from 1 kg PWR-UO_2 fuel at 44 MWd/kg used in a four year refuelling cycle and reprocessed three years after removal from the reactor core. Reprocessing losses were assumed to be 0.2% of U and Pu.

The curve in Figure 1 for HLLW minus minor actinides show that such high-level waste from a modified Purex with complete Np-separation and total Am+Cm removal reaches the uranium reference line after about 400 years.

In order to reach the reference uranium line at about 400 years the requirement put on Np recovery is smaller than on that of Am+Cm recovery, *i.e.* a factor of >2.5 for Np and a factor of >124 for Am+Cm. Both factors seem to be within reach for a well-designed P&T system.

Figure 1: The calculated potential hazard as number of ALI:s from 1 kg PWR-UO$_2$ fuel at 44 MWd/kg used in a four-year refuelling cycle and reprocessed three years after removal from the reactor core

Reprocessing losses were assumed to be 0.2% of U and Pu. MA stands for minor actinides (Np, Am, and Cm)



Flow sheets for P&T of minor actinides

There are in principle two main alternatives to be discussed when designing a flow sheet for P&T of minor actinides:

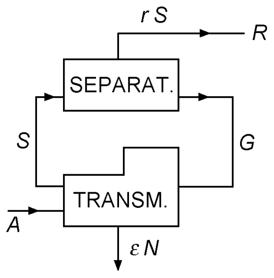
- On-line continuous processing and purification of a continuous target stream. This alternative is often suggested for future systems. It requires a liquid fuel reactor of molten salt or molten metal type, or perhaps target pebbles in a pebble bed reactor.
- Batch-wise processing of irradiated target material. This is the classical way to operate the nuclear fuel cycle and the irradiation target/fuel can in principle be of any type.

We will begin by showing how the losses from a continuous system can be calculated.

Losses from continuous P&T

Figure 2 illustrates a small flows heet for continuous separation/purification of a continuous side stream in a liquid P&T system. The material to be transmuted enters at *A* as a liquid target, flows through the separation/purification circuit where a small amount is lost to the waste stream *R*, which also contains the transmutation products. Then, the purified target flows back into the transmutation device again.

Figure 2: A flow sheet for continuous separation/purification of a side stream in a continuous P&T system using a special transmuter able to handle a liquid target stream



Let us introduce the following symbols and definitions:

- A = rate of addition of target atoms to transmuter;
- N = number of target atoms in transmuter;
- S = Transfer rate to separation process;
- $A = \varepsilon N + rS$; where ε is transmutation rate constant;
- $C = N/V$; where V is irradiated volume;
- $v = kV$ = volume flow rate to separation process;
- $S = vC$; $R = rS$ = loss rate to waste;
- $\varepsilon N = \phi_{avg}\, \sigma_{avg}\, N$; $\varepsilon = \phi_{avg}\, \sigma_{avg}$;
- $Q = A/R$; needed reduction factor;
- $r = \phi_{avg}\, \sigma_{avg}/[k(Q-1)]$, but $\phi_{avg}\, \sigma_{avg}$ is of the magnitude 10^{-8} for fast neutrons.

Example: Permissible Np loss to waste in continuous P&T

Using the estimated reduction factor of about 2.5 for Np we now obtain:

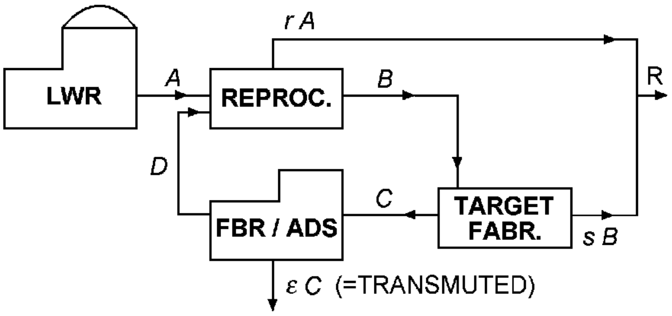
- $\sigma_{avg} \sim 2\ b = 2 \times 10^{-28}\ \mathrm{m}^2$ (in a fast reactor or ADS system);
- $\phi_{avg} \sim 1 \times 10^{19}\ \mathrm{n\ m^{-2}\ s^{-1}}$;
- $Q = 2.5$;
- assume one transmuter volume is purified per day;
- $k = (1/86\,400)\ \mathrm{s}^{-1} = 1.16 \times 10^{-5}\ \mathrm{s}^{-1}$;
- $r = (1 \times 10^{19} \times 2 \times 10^{-28})/(1.16 \times 10^{-5} \times (2.5^{-1})) = 5.75 \times 10^{-5} = 0.00575\%$ of separated amount of Np.

The value of 0.00575% maximum permissible loss now has to be compared to the permissible loss to waste from a batch-wise (discontinuous) P&T process as calculated in the next paragraph.

Losses from batch-wise P&T

Figure 3 shows a possible batch-wise P&T cycle where Np in LWR fuel is separated in a modified Purex process and transferred to a target production facility. The solid Np targets are transferred to a special transmutation facility consisting of *e.g.* a fast reactor or an ADS device. After irradiation, targets are dissolved and added to the Purex feed. The calculation trick is now to follow a given number of initial atoms cycle after cycle until recycling ends, and then to sum losses.

Figure 3: A possible batch-wise P&T cycle for Np from LWR fuel



Let us introduce the following symbols and definitions:

- A = incoming Np amount in a single batch, first cycle;
- $B = (1 - r)A$;
- $C = (1 - s)B = (1 - r)(1 - s)A$;
- $D = (1 - \varepsilon)C = (1 - r)(1 - s)(1 - \varepsilon)A$.

Then for the second cycle of remaining Np from the same batch we can write:

- $B = (1 - r)D = (1 - r)^2(1 - s)(1 - \varepsilon)A$;
- $C = (1 - s)B = (1 - r)^2(1 - s)^2(1 - \varepsilon)A$;
- $D = (1 - \varepsilon)C = (1 - r)^2(1 - s)^2(1 - \varepsilon)^2A$.

Etc.… cycle after cycle. Finally after an infinite number of cycles we have:

$$D = (1 - r)^\infty(1 - s)^\infty(1 - \varepsilon)^\infty A$$

The total amount of Np lost to waste is R, which can be calculated by summing all losses as:

$$R = A(r + r(1 - r)(1 - s)(1 - \varepsilon) + r(1 - r)^2(1 - s)^2(1 - \varepsilon)^2 + \ldots) + \\ A(s(1 - r) + s(1 - r)^2(1 - s)(1 - \varepsilon) + s(1 - r)^3(1 - s)^2(1 - \varepsilon)^2 + \ldots)$$

or:

$$R = Ar\Sigma(1 - r)^n(1 - s)^n(1 - \varepsilon)^n + As(1 - r)\Sigma(1 - r)^n(1 - s)^n(1 - \varepsilon)^n \text{ for } n \text{ cycles}$$

which simplifies to:

$$R = A(r + s - sr)\Sigma(1 - r)^n(1 - s)^n(1 - \varepsilon)^n$$

The values with exponent n are all smaller than one. Using the rules for sums of infinite powers series, this becomes for an infinite number of cycles:

$$R = A(r + s - sr)/(r + s + \varepsilon - rs - r\varepsilon - s\varepsilon + rs\varepsilon)$$

and finally we can write:

$$R/A = (r + s - sr)/(r + s + \varepsilon - rs - r\varepsilon - s\varepsilon + rs\varepsilon) = 1/Q$$

Example: Permissible Np loss to waste in batch-wise P&T

Using the estimated total reduction factor of about 2.5 for Np we now obtain:

$s = 0.0001$ (from MOX-fuel experience), $\varepsilon = 0.05$ (estimated from SUPERFACT expts. [2])

but:

$$Q = A/R = 2.5, \text{ which now yields } r = (Qs - s - \varepsilon + s\varepsilon)/(1 - Q + sQ - s - \varepsilon + s\varepsilon) = 0.032$$

Hence, with these assumptions, a total loss of max 3.2% of Np in one reprocessing cycle is permissible for a reduction of the total Np loss to waste by a factor of 2.5 by P&T. This is much larger than the 0.00575% maximum permissible loss calculated for continuous processing.

Conclusions

The examples given above for Np show that a batch-wise operation often can tolerate a much higher fractional loss than a continuous process when running P&T with the same total loss after an infinite time. For recycling of Am in a batch-wise operation the maximum tolerable cycle loss of Am is 0.0306% in an extended Purex type process. This value must also include the loss to wastes in an Am/Cm separation if Am, but not Cm, is recycled for transmutation.

The intent of this paper was to show how permissible losses can be estimated from the type of flow sheet, the transmuter efficiency, and the acceptable total loss of un-transmuted material to waste.

The calculation of permissible losses forms a connection between the requirements on a transmutation device and on the associated chemical processing plant. The acceptable losses in the separation process can of course be based on any arbitrary requirement regarding the total losses, *e.g.* set by law makers. The use of a hazard index was only chosen here as a simple means to get data on acceptable total losses for illustrative purposes. Furthermore, it is known that the isotopic composition of any element is likely to change during the T-step. To accommodate this in case of batch-wise processing, one has to revert to the equation for R as written in terms of a separate transmutation rate for each recycle and sum the losses cycle by cycle.

Finally we would like to point out that the loss from a continuous process ended after any reasonable time must also include remaining material in the transmuter. This is also true in case of batch-wise P&T operations. Hence the permissible cycle losses are smaller in both these cases in comparison with those calculated assuming infinite operation time.

Acknowledgement

The authors would like to thank Anders Landgren, Ph.D. for valuable discussions regarding different ways to calculate permissible losses.

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Session II

Progress in transmutation fuels and targets

Chairs: D. Warin, Y. Arai

Advanced fuel fabrication processes for transmutation

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Abstract

This paper describes the latest developments and process optimisation at the JRC-ITU for the fabrication of oxide fuels and targets for the transmutation of minor actinides. Concepts considered at ITU have been based on the inert-matrix fuels (IMF) either in homogeneous [e.g. $(An,Y,Zr)O_2$*], or heterogeneous CERMET composite forms [e.g.* $(An,Y,Zr)O_2$ *in molybdenum], where "An" indicates any mixture of Pu, Np, Am. The processes described here have been qualified for the fabrication and characterisation of fuels for their irradiation in the HFR reactor. The experience gained in their fabrication and characterisation is presented. Future process development will focus on U-based fertile fuels in oxide, nitride or carbide form, as will be required for Gen-IV fast reactor systems.*

Introduction

The development of fuels and targets for transmutation of actinides is one of the most important issues in the proposed P&T concepts. Different requirements for the fuel and target design can be defined, depending on the fuel cycle strategy. For example, with multiple recycling and use of fast reactors, high fissile contents and the possibility to reprocess the fuel are of key importance. In contrast, in a once-through transmutation scenario, the stability of the material at ultra-high burn-up and as spent material for geological disposal is crucial. If the extent of transmutation must be as high as possible, the development of inert-matrix fuels (IMF) is a promising approach. Thus, uranium-free fuels could burn plutonium and minor actinides (MA) more efficiently since their activation source (^{238}U) is eliminated. As the transmutation process requires fissioning of actinides, a high burn-up of the fuel/target is required to reach high efficiency. This will lead to high demands on the performance of the fuel in view of the concomitant high fission gas and helium production, fuel swelling, and chemical and mechanical fuel cladding interaction. All of these will exceed the levels known in existing fast reactor fuels. Solid solutions and dispersions of actinide oxides in a ceramic (cercer) or metallic (cermet) uranium-free matrix are considered as targets for the transmutation of americium. The main advantage of composite targets compared to solid solutions lies in the potentially minimised irradiation-induced property changes in the fuel, achieved by localising the fission damage in a limited geometric domain within the fuel. Cermet fuels offer further advantages such as their ability to retain fission gases, volatile fission products and helium in the fuel, and their high thermal conductivity, which either permits higher actinide loading or lower operating temperatures. The latter thereby increases safety margins under normal and off-normal reactor conditions.

The performance of these innovative fuels, which have a direct impact on the reactor design and transmutation efficiency, is largely unknown, due to the lack of knowledge of their in-pile behaviour (*e.g.* swelling, gas release, restructuring, volatile species redistribution, corrosion) and their main thermal, chemical and mechanical properties. At ITU, the main efforts so far have concentrated on developing and optimising advanced fabrication processes for these fuels and their characterisation needed to satisfy reactor design and safety issues for their qualification, and ultimately to enable a detailed understanding and prediction of their performance under irradiation.

Fabrication of targets for transmutation and incineration of minor actinides requires more stringent radiation protection measures than currently necessary for the manufacture of conventional UO_2 and MOX fuels. Whereas MOX can be fabricated in glove boxes, the handling of minor actinides requires extra shielding in the form of lead (for gamma radiation) and water (for neutron radiation), and remote operation. As a result, process simplification and automation are necessary. In addition, the waste generated should be minimised. The fabrication procedure should not generate dust, which could collect on the surfaces of the glove boxes and the equipment therein, so that operator intervention is facilitated and radiation exposure to the personnel minimised. For these reasons, dust-free fabrication has become a major development topic at the ITU where a hybrid process consisting of a combination of sol-gel [1,2] and porous bead infiltration techniques [3] are being developed. Blending of the particles and the matrix powder by conventional methods is required for the composite fuel type. Process development continues on a laboratory scale, but keeping industrialisation as the long-term goal.

In this paper, a detailed description of the latest developments on the fabrication process is given. In addition, some of the results obtained for the fabrication of fuels by optimised processes will be compared with similar fuels fabricated by the standard ITU infiltration process.

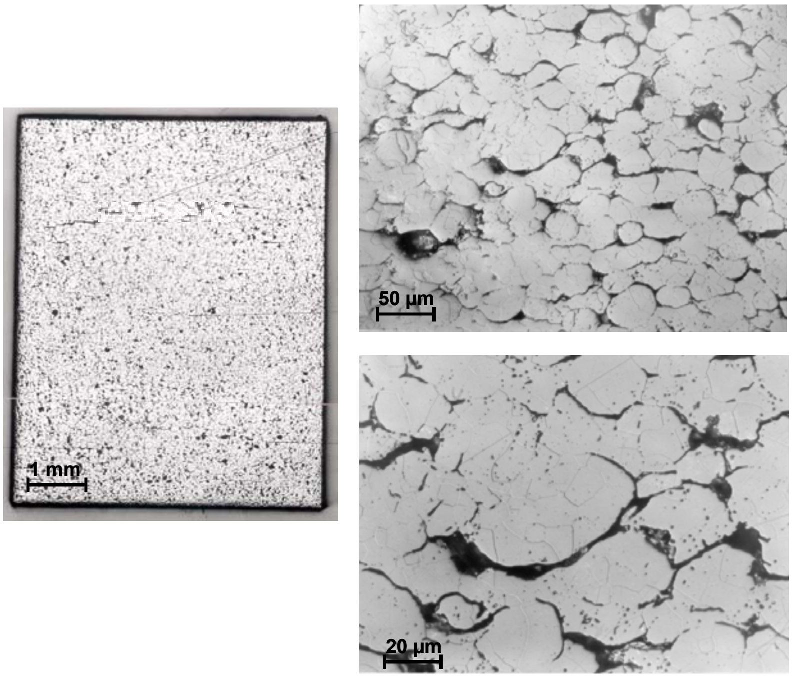
Experimental procedure

The external gelation method is a sol-gel process that has been widely used for the production of nuclear fuels for high-temperature reactors (HTR) in Germany and the USA and more recently in Japan and China. For this purpose it is well suited, as the final product is a fuel or fertile kernel, with a well-specified diameter, which depends on the fissile content in the particle. At the ITU [1,2], the same droplet to particle conversion step is produced to produce small polydisperse particles, from which pellets can be obtained by compacting them to form pellets. Sintering thereof gives the product pellets.

Powders produced in this way have inherent advantages. In particular the liquid to solid conversion step is essentially a dust-free route, which is extremely important for facilities in which Am or dirty (high ^{241}Am content) Pu is handled. In addition, the process yields a completely atomically homogeneous product in the form of a solid solution of the components. Present industrial production of MOX by the MIMAS process strives for homogeneity, but cannot due to its nature be as successful as the liquid processing route. A major disadvantage in liquid processing routes, however, is the reduction in margins of criticality compared to powders, which can limit throughput and flexibility in blending significant quantities for Pu with different isotopic vectors in large industrial installations.

The direct pelletisation step at the ITU gives special emphasis to the production of relatively small particles, in the 20-150 μm range, to achieve easy pressing and a good microstructure. The microstructure of the final pellets produced from these particles pores surrounding the precursor particles and is often known as the “blackberry structure”. Mixed experience has been made at the ITU using the direct pelletising process. While U-MOX pellets with Pu contents between 4 and 20% sinter extremely well and often result in pellet densities close to 98% of the theoretical density (TD), other materials do not sinter well and low densities with the blackberry structure are found. An example is shown in Figure 1 for a $Pu_{0.090}Y_{0.153}Zr_{0.757}O_2$ fuel pellet.

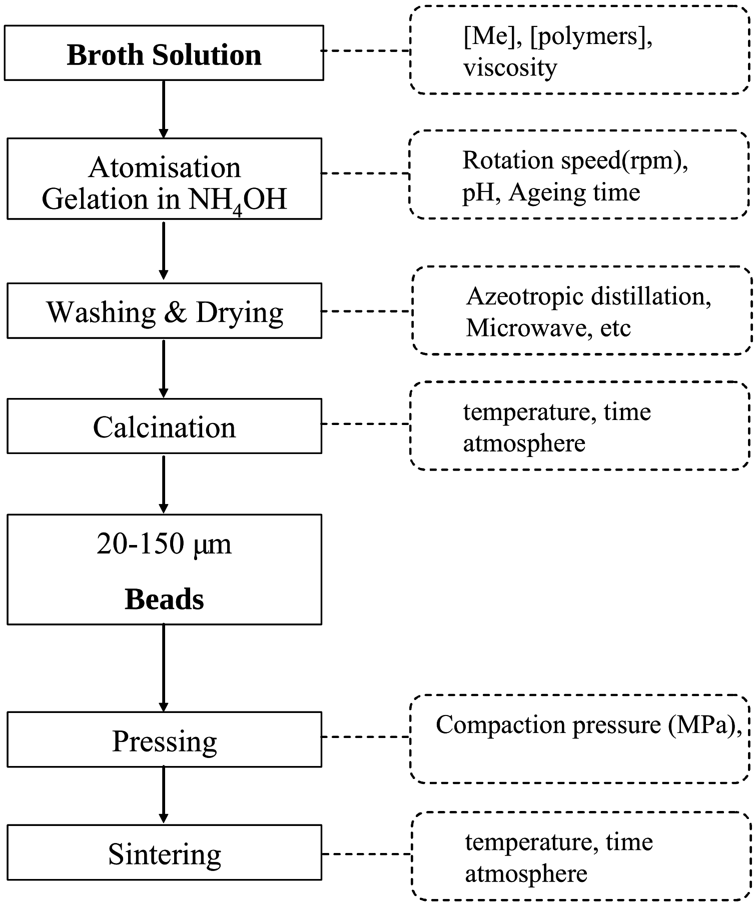
Figure 1: Optical micrographs of an axial section of a $Pu_{0.090}Y_{0.153}Zr_{0.757}O_2$ pellet showing the “blackberry” microstructure



Within the external gelation process, shown in Figure 2, there are a large number of parameters which can be varied (*e.g.* metal and polymer concentration, polymer type, ageing time, etc.) so that an optimisation of the parameters for each material and even different composition ratios thereof can be extremely time consuming. Therefore a process improving the consistency of the manufacturing, as well as yielding the required microstructure and pellet density, is sought.

Tests have shown that it is possible to compact the sol-gel beads and granulate them in a hammer mill before pressing the subsequent powder to give a perfect product. This solution not only lacks elegance, but also results in fine powders, which must be avoided especially when handling minor actinides. The possibility to improve the microstructure and density of the final fuel pellets by tailoring the particle porosity has also been investigated and tested as an option in the process optimisation. In this case, carbon black is dispersed as a powder in the initial feed solution before the droplet dispersion. This additional carbon acts as a pore former and softens the kernels and improves their compaction characteristics. Simultaneously, the pores created within each precursor particle can be retained to some extent during sintering to give the desired microstructure. In particular the sintering between individual precursor particles is enhanced in comparison to their own internal sintering, so that that the “blackberry” structure is diminished.

Figure 2: The sol-gel external gelation process and its parameters



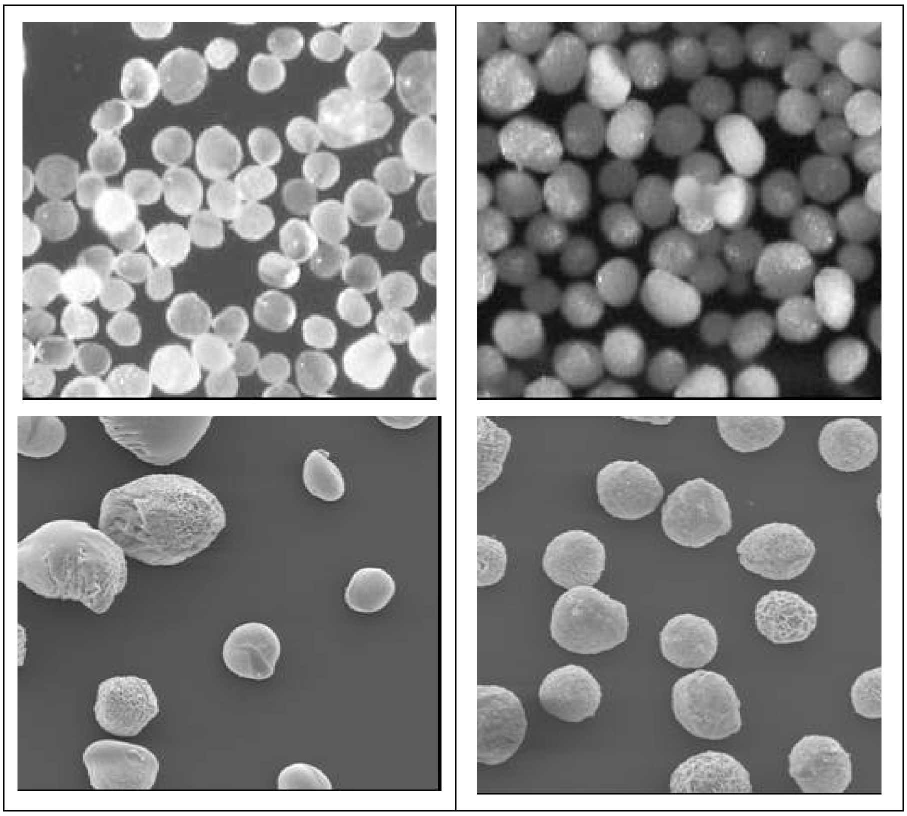
The increased porosity of the calcined particles is an excellent attribute for the infiltration process developed at ITU for the production of samples containing minor actinides [3]. This process is based on the infiltration of porous beads, produced by a sol-gel process, by the actinide nitrate solution. Following evaporation of the water and conversion of the infiltrated actinide nitrate to oxide in a calcination step, the beads are compacted and subsequently sintered to give the final product pellets.

Results and discussion

Yttria-stabilised zirconia (YSZ) beads have been prepared with and without carbon in the feed solution. Their form is compared using optical and secondary electron microscopy (SEM) in Figure 3. The distribution in size is similar in both cases. Most importantly, however, is the shiny, almost glassy, nature of the beads produced without carbon addition. In contrast, the beads produced with carbon in the feed solution have less well-defined and more open structures. The SEM measurements (see lower panel in Figure 3) confirm this general effect, but it is also difficult to quantify. The open structure is clearly visible in the beads produced with 20 g/l carbon in the feed solution. It is also present in some of the beads produced without carbon, but here there are also larger numbers of beads with perfectly smooth surfaces.

The calcined YSZ beads have been infiltrated with americium nitrate solution and were again thermally treated in air to convert the Am nitrate to oxide, to give an intermediate product consisting of YSZ + AmO_2. These composite beads, though not sintered maintain an excellent flowable character and resistance to abrasion and loss of fine particles of either AmO_2 or YSZ.

Figure 3: Optical and SEM pictures of YSZ beads produced without (left) and with (right) carbon addition (20 g/L) to the sol-gel feed solution



The infiltrated beads were compacted in a biaxial press. Increasing the carbon content of the feed solution gives pellets with increasing final density. Without carbon addition the density is 90.0±1.0% TD and increases to 92.6±0.4% TD when the feed solution contained 20 g/l of carbon. This increase in density is an indication that the particles are easier to compact, *i.e.* their crush strength is reduced on addition of carbon. This can be reconciled with the increased pore structure of the precursor beads achieved with carbon addition. Pictures of pellets produced with and without carbon addition are shown in Figure 4, while their microstructure is shown in Figure 5. A dramatic improvement is observed. Modification of the process conditions by the addition of carbon to the feed solution eliminates the blackberry structure (a cause of the lower density) and provides the pellet with a more uniform microstructure.

Figure 4: Visual aspect of homogeneous HELIOS pellets

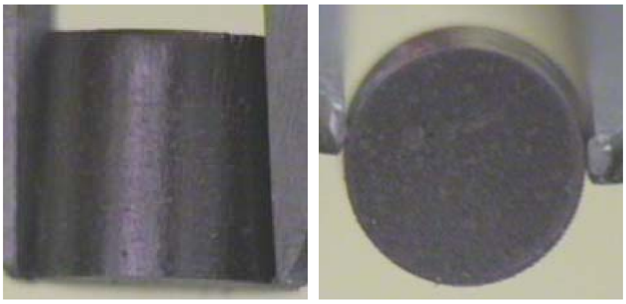
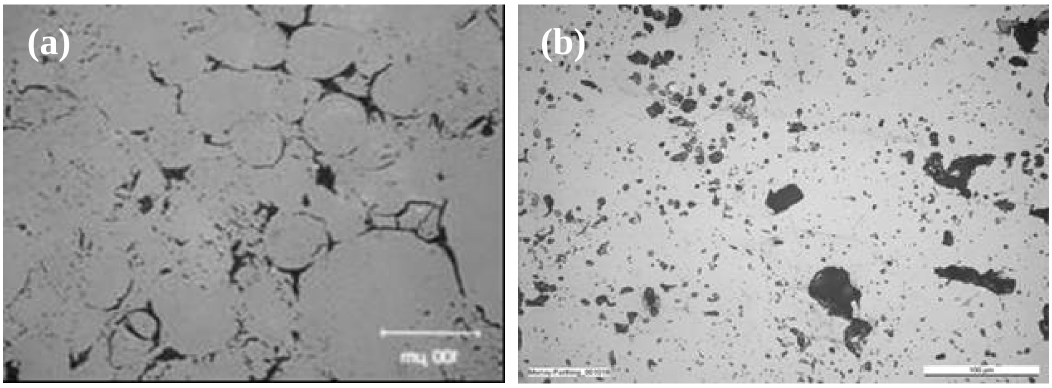


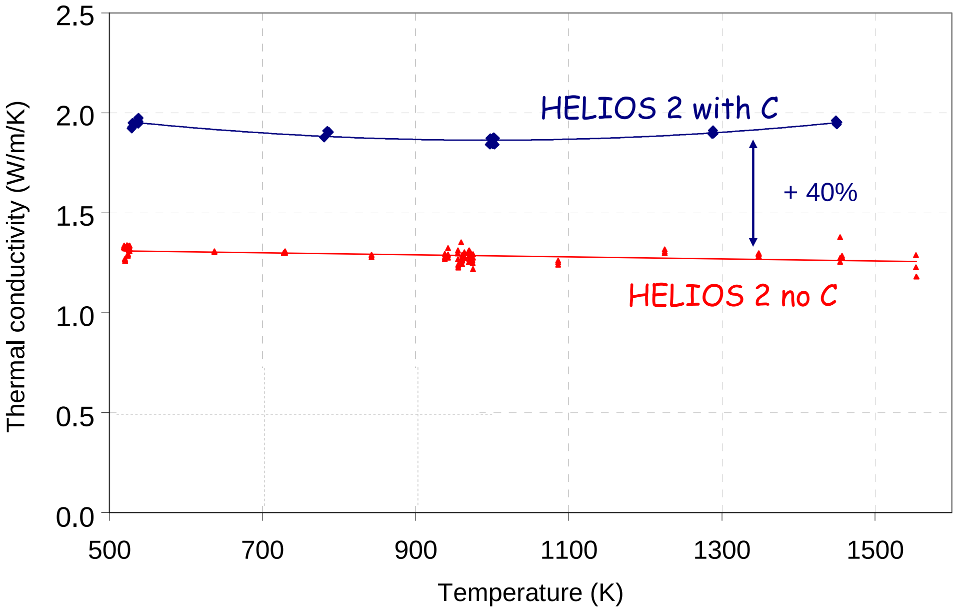
Figure 5: Microstructure of ZrYAmO2 pellets fabricated (a) with no C addition and (b) with C addition (HELIOS 2)



The thermal conductivity of the fuel is one of the most important properties in the licensing of a fuel for a qualification test. The thermal diffusivity, α, of the highly γ-active samples is measured in a shielded "laser-flash" device [4], and the heat capacity determined by differential scanning calorimetry (DSC) in commercial equipment (Netsch 409). The thermal conductivity $\lambda(T)$ is then calculated from the independent measurement of the thermal diffusivity, $\alpha(T)$, the heat capacity, $C_p(T)$, and corrected for the density $\rho(T)$.

Several samples containing americium have been measured, including homogenous ($Zr_{0.78}Y_{0.16}Am_{0.06}O_{1.92}$) fabricated both by the conventional process (C-free oxide fuel) and by the addition of C to the sol-gel broth solution (C-added oxide fuel). Thermal annealing cycles with increasing maximum temperature were performed, but no effect of recovery of radiation damage was observed in the thermal diffusivity. Although the two samples have the same chemical composition, their thermal conductivity is significantly different (see Figure 6). The thermal conductivity of both samples lies within the range typical for this type of zirconia-based compounds. Nevertheless, the addition of C to the broth for the production of the YSZ precursor beads not only improves the microstructure of the fuel, but also induces a remarkably significant increase (40%) of the fuel thermal conductivity.

Figure 6: Thermal conductivity of ($Zr_{0.78}Y_{0.16}Am_{0.06}O_{1.92}$) pellets fabricated (a) with no C addition and (b) with C addition (HELIOS 2)



Conclusions

Addition of carbon to the feed solution in the sol-gel external gelation process provides a means to improve the microstructure of the pellets and eliminate the "blackberry" structure often found in directly pressed sol-gel beads. This is a particular problem for large (500 μm) particles, and has often been observed at ITU even where more favourable polydisperse (20-150 μm) particles are prepared. It seems that the addition of 10-20 g/l of carbon to the feed solution is sufficient to improve the microstructure and obtain higher density pellets. The effect is largely due to the decreased crush strength of the beads permitting higher green densities of the pellets which, coupled with the increased porosity of the precursor beads, enables a more uniform sintering to give a more homogeneous microstructure. In particular, carbon addition reduces internal sintering of the beads to eliminate the "blackberry" microstructure. The addition of carbon has a further beneficial effect. Namely, the porosity increase permits infiltration of higher quantities of actinide, thereby giving the infiltration technique greater flexibility.

The good thermal properties of cermet compounds (not discussed in this publication) is attractive from the pin design point of view, although the actual in-pile behaviour will have to be determined, in particular to consider processes such as the material swelling under fast neutron irradiation. Complementary irradiation programmes, carried out within the European Integrated Project EUROTRANS, in the PHENIX sodium fast reactor in Marcoule (France), and in the HFR material testing reactor in Petten (Netherlands) are ongoing. The main goals of these experiments are the demonstration of the in-pile thermo-mechanical and chemical stability of the fuel, the fission gas and helium accumulation and release behaviour, and the comparison of this with other fuel types such as cercer oxides, nitrides and metallic fuels.

Acknowledgements

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Development status of transuranic-bearing metal fuels

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Abstract

Within the framework of the Global Nuclear Energy Partnership/Advanced Fuel Cycle Initiative programme, the United States Department of Energy is conducting research and development, and performing demonstration activities on transmutation fuels. Transmutation fuels contain transuranic elements (Pu, Np, Am, Cm) in various proportions recovered from reprocessing of spent nuclear fuel. Along with ceramic fuel forms with an emphasis on oxide fuels, the US programme is focusing on metallic fuel for transmutation applications.

This paper provides a summary of the available data to date along with a report on the research and development and demonstration programme that are in place to achieve the qualification of MA-bearing metal fuels for use in sodium-cooled fast reactors. The development programme is gauged via the use of the technology readiness levels, a specific description of which is also included in the paper.

1 Introduction

Under the Advanced Fuel Cycle Initiative programme, the United States Department of Energy is conducting research, development and demonstration activities on transmutation fuels. Transmutation fuels contain transuranic elements (Pu, Np, Am, Cm) in various proportions recovered from reprocessing of spent nuclear fuel. Along with ceramic fuel forms with emphasis on oxide fuels, the US programme is focusing on metallic fuel for transmutation applications.

The US has considerable experience with the use of metal fuel, especially concerning experimental fast reactors (EBR-I and EBR-II). The early applications were based on the binary (U-Zr) metal fuels; considerable maturity with the ternary fuel (U-Pu-Zr) was established during the Integral Fast Reactor (IFR) programme between 1984 and 1994. Hundreds of ternary fuel rods were irradiated in EBR-II and FFTF using a variety of clad materials. At the termination of the programme, sufficient steady-state and transient performance data had been collected to qualify the ternary fuel for operation in a sodium-cooled fast reactor up to 10% burn-up.

Transmutation fuels are quite different than conventional fuels because of the following issues:

- multiple elements in the fuel (U, Pu, Np, Am, Cm);
- varying thermodynamic properties (*e.g.* high vapour pressure of Am);
- impurities from separation process (*e.g.* high lanthanide carryover with some separation processes);
- high burn-up requirements;
- high helium production during irradiation;
- remote fabrication and quality control requirements;
- fuel must be qualified for a variable range of composition because of:
  - age and burn-up of LWR SNF,
  - introduction of first tier recycle in LWR;
  - changes through multiple passes in FR;
  - variable conversion ratio for FR.

2 Review of metal fuel experience

An extensive review of historical US conventional fast reactor fuel technology, with an expanded discussion of the above, is provided by Crawford, *et al.* [1]. Conventional fuel is defined as fuel forms that use highly-enriched uranium (HEU) or U-Pu that have been developed in domestic and international fast reactor programmes. Features believed to be pertinent to transmutation fuel technology are summarised, with the reader directed to the appropriate literature reference for further detail.

Typically, the fuel portion of the metallic fuel element (the fissile component of metallic fuel) is uranium, uranium-plutonium, or a mixture of uranium-plutonium and the minor actinides (MA). The fuel alloy is stabilised using, typically, a 10-30% addition of zirconium to both increase the melting point and minimise fuel-cladding chemical interaction (FCCI). The fuel slug is thermally bonded to the cladding using sodium. The sodium provides a very high thermal conductivity medium by which heat is easily transferred to the cladding and reactor coolant. A fission gas collection plenum is provided to capture the release fission product gases. The fuel and sodium have typically been sealed inside a stainless steel cladding of austenitic or ferritic-martensitic (FM) composition or nickel-based alloy. The current designs employ the low swelling FM stainless steels. Future designs may employ more advanced clad materials to achieve burn-up levels exceeding 20%.

A summary of the metal fuel experience is provided in Table 1.

Figure 1

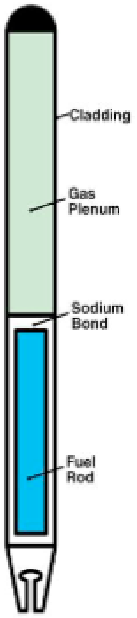


Table 1: Summary of conventional metal fuel experience

Campaign	Mark-I/IA	Mark-II/IIC/IICS	Mark-III/IIIA	Mark-IV	Mark-V/VA*
Fuel alloy (wt%)	U-5Fs	U-5Fs U-10Zr	U-10Zr	U-10Zr	U-20Pu-10Zr
^{235}U enrichment (%)	52	67-78	66.9	69.6	Variable
Slug diameter (mm)	3.66	3.30	4.39	4.27	4.27-4.39
Smeared density (%)	85	75	75	75	75
Burn up limit (at.%)	2.6	8.9	10	N/A	TBD
Plenum to fuel volume ratio	0.18	0.68-1.01	1.45	1.45	1.45
Cladding material	SS 304L	SS304L SS 316	CW 316 CW D9	HT 9	HT 9 CW 316

* Conversion to the Mark-V/VA fuel types was not started before EBR-II was terminally shut down in 1994.

3 Transmutation applications for metal fuel

Until the beginning of the AFCI programme, there was limited experience with MA-bearing metal fuels. However, it was decided that metal fuel provides the potential for excellent performance as a MA-bearing fuel, considering the demonstrated performance of conventional metal fuels. Study of the following issues forms the basis for defining the current focus areas of metal MA-bearing fuel research and development:

- Demonstration of MA-bearing oxide feedstock reduction to metal alloy feedstock.
- Due to Am metal volatility, fabrication with high Am retention must be demonstrated, using revised casting technology utilising higher pressure systems, shorter heating times and removing conditions that would promote Am vapour deposition.
- Demonstration of metal fuel fabrication with low irretrievable losses.
- Metal fuel properties must not be seriously degraded as compared to the U, Pu, Zr system performance by the addition of the MA (Am, Np and Cm).

- Demonstration of an acceptable level of fuel-cladding-chemical-interaction (FCCI) with fuel that includes rare-earth impurities and MA fuel constituents over the lifetime of the fuel up to its burn-up limit, ~20 at.%.
- Assuming fuel melting and FCCI characteristics are acceptable, behaviour of MA-bearing fuel in over-power transients and run beyond cladding breach should be acceptable as well.
- Burn-up limitation extensions to about 238 MWd/kgM (at $39 \times 1\,022$ n/cm^2 or approximately 200 dpa) can be anticipated with current ferritic/martensitic steel cladding. Up to 300 MWd/kg may be achievable with increased high-temperature cladding strength.

An important question related to the use of U-Pu-Am-Np-Cm-Zr alloys as transmutation fuels are the unknown phase equilibria in the multi-component alloy system. The potential for immiscibility and formation of an inhomogeneous microstructure is not a fuel performance issue, as shown by the excellent performance of multi-phase U-Pu-Zr fuel. Rather, the formation of low melting phases in the complex alloy system is an issue that must be experimentally determined. Recent experimental irradiations and out-of-pile studies conducted as part of the Advanced Fuel Cycle Initiative (AFCI) programme indicate that this is not likely to be a serious issue.

4 Irradiation testing with MA-bearing metal fuels

A number of steady-state transuranic metal fuel tests have been conducted: EBRII-X501, AFC1 series conducted in the Advanced Test Reactor (ATR) by thermal flux filtering with cadmium, and METAPHIX-1 &-2 conducted in the Phénix reactor for CRIEPI Japan. Current irradiation experiments include the AFC2 series in the ATR, METAPHIX-3 and the FUTURIX-FTA series, both conducted in the fast spectrum Phénix test reactor.

Varying amounts of U, Pu, Am, Np in Zr matrix have been tested in ATR up to burn-up levels as high as 40%. In addition to fertile fuels (containing U), some non-fertile fuels were also tested (uranium-free) in the early part of the programme. Even though most of the US-based irradiations of MA-bearing metal fuels were conducted in the ATR with thermal neutron filtering, the results to date indicate that for critical performance parameters the data correlate very well with earlier ternary fuel experiments in fast reactors. Figure 2 shows the effect of cadmium filtering on the neutron spectrum in the ATR testing position.

5 Fabrication of MA-bearing metal fuels

All the samples prepared for irradiation experiments were fabricated using an arc-casting method, which provides quick melting and casting with minimal Am losses, but it is not amenable to the large-scale production that is needed. The advanced casting methodology that currently being worked on addresses the following issues:

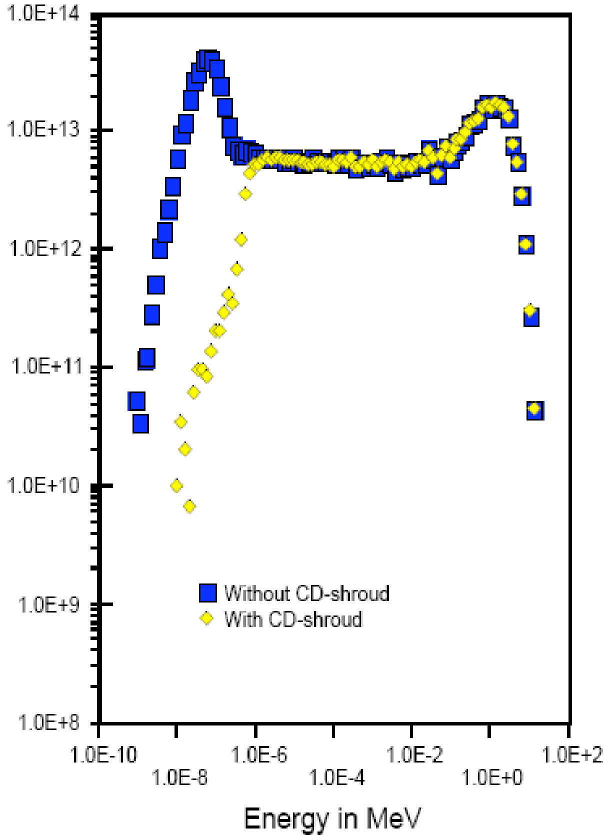
- retention volatile elements (Am);
- application for hot cell use;
- increase in melt usage;
- reduction of materials losses and waste production.

The developmental approach that is taken includes:

- bench-scale system (BCS);
- engineering-scale system (ECS);
- advanced casting system (ACS).

The BCS is currently being used to test Am volatility issues and different mould techniques to minimise losses.

Figure 2: Effect of cadmium filtering on the neutron spectrum in the ATR testing position



6 Summary and conclusions

Metal fuel development for transmutation applications continues. Considerable characterisation activities include:

- measurement of thermodynamic properties;
- phase diagrams/phase distribution/microstructure;
- diffusion-couple measurements.

Also, there is considerable effort in looking at cladding development in the following areas:

- high-fluence data on HT-9 (200 dpa);
- looking at advanced options (ODS);
- considering lined/coated cladding options.

We believe the efforts to date have demonstrated the technical feasibility of MA-bearing metal fuels (>10% burn-up) Advanced modelling and simulation is being used to cover the range of compositions that are of interest as an integral part of the programme.

The engineering feasibility of large-scale fabrication will require additional work, and casting simulations are being used to optimise the fabrication processes.

Acknowledgements

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Design, development and qualification of advanced fuels for an industrial accelerator-driven system prototype

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Abstract

Uranium-free fuels for minor actinide burning in a 400 MWth accelerator-driven system are under development within the EUROTRANS European Integrated Project. The present paper gives an overview of the results for the ongoing work.

Two kinds of fuels have been selected as primary candidates: they are composite materials consisting of $(Pu_y,MA_{1-y})O_{2-x}$ *particles dispersed in a molybdenum or magnesia matrix. The performances of these MgO-CERCER and Mo-CERMET fuels have been modelled under normal operation as well as under transient and accident conditions. Three zone cores are required to obtain reasonable operational and safety parameters that satisfy the general and specific requirements of the overall design for the lead-cooled European Facility for Industrial Transmutation.*

In parallel to design and safety studies, in-pile and out-of-pile experiments are carried out to gain knowledge on the fuels. Irradiations investigate the helium release mechanisms (BODEX experiment in HFR), the role of microstructure and temperature on gas release and fuel swelling (HELIOS experiment in HFR) and the behaviour of the fuels in ADS representative flux conditions (FUTURIX-FTA in PHENIX). The out-of-pile experiments consist in measuring thermal and mechanical properties of the fuels, and assessing the chemical compatibility between fissile phases, matrices, clad and coolant, in order to supply relevant and accurate properties for fuel design, fuel performance and safety modelling.

Introduction

Fuel to be used in an accelerator-driven system (ADS) for transmutation in a fast spectrum can be described as highly innovative in comparison with fuel used in a critical core. ADS fuel is not fertile, *i.e.* there is no uranium, so as to improve the transmutation performance. It necessarily contains a high concentration of minor actinides (about 45% to 70%) and plutonium (30% to 55%) whose isotopic vector typically consists of 80% to 90% of even isotopes ^{238}Pu, ^{240}Pu and ^{242}Pu, 10% to 20% of odd isotopes ^{239}Pu and ^{241}Pu. This unusual fuel composition results in high gamma and neutron emissions during its fabrication, as well as constraints on core operations. This is due to: *i)* thermal and thermodynamic properties of the minor actinide (MA) compounds, which are poorer than those of major actinides; *ii)* volatility of the americium species; *iii)* significant production of helium during irradiation. Thus, an optimal ADS fuel must strike the best compromise between: *i)* constraints of a thermal nature (margins in relation to melting or decomposition/vaporisation) or of a mechanical nature (cladding failure through fuel-cladding interactions and internal pressure); *ii)* the chemistry of the fuel and its cladding (cladding corrosion, fuel-cladding eutectics); *iii)* neutron- and technology-related constraints associated with subcritical cores (themselves defined by the core's level of subcriticality and the power of both the accelerator and the spallation target).

CERCER and CERMET composite fuels, consisting of particles of $(Pu,MA)O_2$ phases dispersed respectively in matrices of magnesia and molybdenum, are under investigation within the frame of the ongoing European Integrated Project EUROTRANS (European Research Programme for Transmutation), which aims at performing a conceptual design of a 400 MWth transmuter: the European Facility for Industrial Transmutation (EFIT) [1].

The performance and safety of EFIT cores loaded with CERCER and CERMET fuels are under evaluation. Out-of-pile and in-pile experiments are carried out to gain knowledge on the properties and the behaviour of these fuels. The current paper gives an overview of the progress of work.

Core design and fuel performance

The EFIT core is being designed for a thermal power of about 400 MWth, aiming at: high transmutation efficiency, low beam current requirement, flat power distribution, low reactivity swing during burn-up, low core pressure drop as well as safety requirements in order to prevent accidents.

The reference working hypotheses have been as follows [2]:

- A proton beam of 800 MeV and 20 mA impinges on a windowless lead target (deposited thermal power of 11.2 MW_{th} and diameter of 78.2 cm), providing the neutron source for the subcritical system with an effective neutron multiplication coefficient (k_{eff}) of 0.97.
- The inlet and outlet temperatures of the lead coolant in the ADS core are respectively 400 and 480°C. The maximum coolant velocity is about $1 m.s^{-1}$, to limit the corrosion and the pressure drop through the core.
- The structural material is ferritic-martensitic steel T91, for which the maximum permitted clad temperature under normal operating conditions is 823 K and the maximum neutronic damage is 90 dpa.
- The isotopic vectors for the transuranium elements (TRU) are determined by mixing the minor actinides arising from spent UO_2 and MOX fuels (45 MWd/kgHM burn-up in a typical PWR) in the ratio 90%:10%.
- In subcritical reactors for burning minor actinides, most fissions should be those of the minor actinides. The maximum destruction of MA is a balance of -42 kg/TWh_{th} (as the fission energy is about 200 MeV/fission) while the Pu content remains constant in the cycle (Pu balance ~0 kg/TWh_{th}).
- Fuel limit temperatures in various accident categories have been specified according to a very conservative approach [3] due to the limited amount of data available. The temperatures (Table 1) are lower for MgO-CERCER than for Mo-CERMET because of expected MgO vaporisation above 1 800K.

Table 1: Categorisation of fuel limiting conditions for beginning-of-life fuels [4]

	MgO-CERCER	Mo-CERCER
Design basis condition categories:		
I	1 750 K	2 300 K
II	1 850 K	2 350 K
III & IV	1 950 K	2 400 K
Design extension conditions	2 150 K	2 450 K

Several design options for both CERCER and CERMET cores have been investigated. The current best designs have both an active core height of 90 cm, three radial zones, the same subassembly hexagonal geometry and size (inner width 178 mm, wrapper width 4 mm and pitch 191 mm), a Pu/MA atomic ratio about 46/54. The differences between the two cores are the matrix fraction in the fuel, the pin diameter and the subassembly layout in the core.

- The CERCER core [2] consists of 42, 66 and 72 subassemblies respectively in the inner, intermediate and outer zones. The fuel pellet diameter is 7.1 mm in the inner and middle zones, and increases to 8.0 mm in the outer zone. The MgO fraction in the fuel is 50% in the inner zone and increases to 57% in the other two zones.
- The CERMET core consists of 42, 90 and 84 subassemblies respectively in the inner, intermediate and outer zones. The fuel pellet diameters are the same for the three zones (8.0 mm). The matrix fraction in the fuel is 65% in the inner zone, decreasing to 57% in the middle and to 50% in the outer zone. The molybdenum isotopic vector is enriched with ^{92}Mo (93%), in order to decrease the neutronic penalties due to ^{95}Mo.

The major pellet, pin and assembly characteristics are given in Table 2. The results of the neutronic modelling performed with the MCNPX code for the CERCER core [2] and the SIMMER-III code for the CERMET core [5] are presented in Table 3.

In both cases:

- The core total power form factor (ratio between peak power density in the fuel to core mean power density in the fuel) meets the specifications (less than 1.7) with 1.55 in the CERMET core and 1.57 in the CERCER core.
- The minor actinide transmutation rates are close to the optimum: 40-43 kgMA/TWh$_{th}$ for both cores with a minor production or consumption of Plutonium (~1-2 kgPu/TWh$_{th}$) after the first three-year cycle of the fuels in the core (+ three years cooling for CERMET fuel).

The effect of irradiation on the thermal, mechanical and thermo-chemical properties is poorly known for these innovative fuels. However, initial thermo-mechanical calculations (TRAFIC and FEMALE simulations) have been performed at BOL, with a simplified description of these heterogeneous fuels and with assumptions concerning the behaviour of the fission gas in the pellets. The fuel and clad maximum temperatures and the maximum clad stress (Table 4) point out differences between the results given by the two codes. Nevertheless, in both cases the temperature and stress values are below the adopted limits, with larger margins for the CERMET fuel.

Table 2: Characteristics of pellet, pin and assembly characteristics for the MgO-CERCER and ^{92}Mo-CERMET reference cores

	MgO-CERCER core			^{92}Mo-CERMET core		
	Inner	Intermediate	Outer	Inner	Intermediate	Outer
Pu/MA	45.7/54.3% (at)			46/54% (at)		
Fuel/matrix	57/43	50/50		35/65	43/57	50/50
Fuel pellet diameter	7.10 mm		8.00 mm	8.00 mm		
Clad inner diameter	7.42 mm		8.32 mm	8.32 mm		
Clad width	600 μm			600 μm		
Fuel pin pitch	13.63 mm		13.54 mm	13.54 mm		
Pin number/assembly	169			169		
Assembly number/zone	42	66	72	42	90	84

Table 3: Neutronic and thermal data for the MgO-CERCER and ^{92}Mo-CERMET cores

	MgO-CERCER core	^{92}Mo-CERMET core
Thermal power (MW)	384	400
k_{eff} at BOL	0.97403	0.97336
ks	0.9511	0.93230
Proton beam current (mA)	13.8	13.8
Active core void worth (pcm)	6 684	7 335
Beta effective (pcm)	148	192
Doppler constant (pcm)	–	-68

Zones	Inner	Middle	Outer	Inner	Middle	Outer
Thermal power (MW)	100	149	150	86	179	134
Average power density ($W.cm^{-3}$):						
– fuel at BOL	399	378	275	270	262	211
– zone at BOL	84	80	74	72	70	56
Hottest pin (at BOL):						
– power (kW)	18	15	17	15	13	12
– average linear power ($W.cm^{-1}$)	205	170	184	162	147	131
Peak pellet linear power ($W.cm^{-1}$)	232	198	218	190	172	154

BOL: Beginning of life.

Table 4: Peak parameter ranges of the hottest CERCER and CERMET rods calculated with FEMALE and TRAFIC codes at BOL (24 hours after start)

	CERCER core	CERMET core
Peak fuel temperature (K)	1 600-1 745	1 100-1 170
Peak clad temperature (K)	790-820	790-795
Maximum stress in the clad (MPa) (limit 127 MPa)	13-23	10-18

Fuel safety assessment

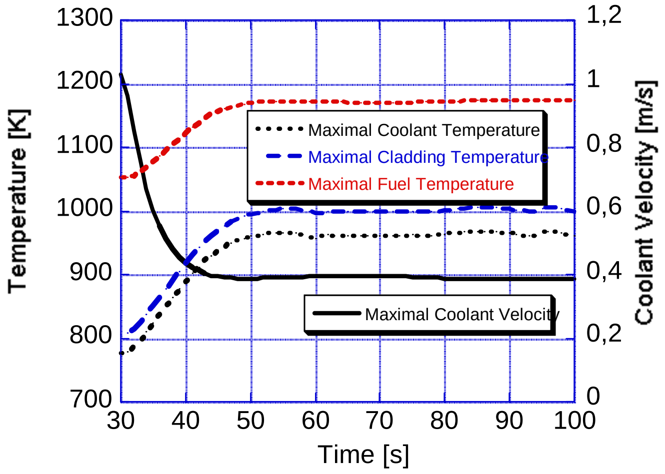
The following transient calculations have been performed to check the compatibility of the preliminary and reference designs with the fuel, clad and coolant safety requirements [3-8]: unprotected loss of flow (ULOF), unprotected loss of power (UTOP), unprotected blockage accident (UBA).

ULOF is a typical transient for demonstrating the impact of power density and core/system design on fuel, clad and coolant temperatures. ULOF simulations by the SIMMER code with the reference CERMET core design [8] point out (Figure 1) that both fuel and clad failure limits are respected. Moreover, the fuel has very large safety margins.

The SIMMER simulation of an UTOP doubling the beam power has not shown any short-term CERMET fuel failures [5].

In the UBA, the effect of the local release of helium after pin failure has to be assessed. Although the high boiling point of the lead coolant prevents coolant boiling, local voiding can be caused by a helium blow-down from the gas plena. A mitigating effect could be the sweep-out of fuel particles. Simulations by the SIMMER code with the reference CERMET core design have shown that pin failure would lead to a local voiding and reactivity addition but the fuel sweep-out effect would lead to a power reduction and would limit the core degradation. The beam should finally be shut off to achieve stable post-accident heat removal conditions [5].

Figure 1: Coolant velocity and temperatures at the upper parts of hottest CERMET fuel subassembly in the case of ULOF [8]



Irradiation tests

As the behaviour of TRU fuel under irradiation is quite unknown, an irradiation programme investigates:

- the irradiation effects on both CERCER and CERMET fuels in EFIT representative conditions (FUTURIX-FTA test [9] in progress in PHENIX reactor);
- the helium behaviour versus temperature and microstructure (HELIOS test to be performed in HFR reactor) [10];
- the helium build-up and release mechanisms versus temperature in ^{10}B (as Am surrogate) doped fuel matrices (BODEX test [11] achieved in HFR reactor);

In addition, post-irradiation examinations of nitride fuels irradiated in the HFR reactor as part of the 5th European Framework Programme CONFIRM project [12], are performed within EUROTRANS project too, as nitride fuels constitute a backup solution for actinide burning. The first results will be available soon.

The composition of the pellets for FUTURIX-FTA and HELIOS experiments are given in Table 5 as well as estimated maximum temperatures in the pellets under irradiation conditions.

Table 5: Composition of the pellets and maximum central temperatures under irradiation conditions

FUTURIX-FTA	
Composition	**T_{max} (K)**
$Pu_{0.80}Am_{0.20}O_{2-x}$ + 86 vol.%Mo	1 865
$Pu_{0.23}Am_{0.24}Zr_{0.53}O_{2-x}$ + 60 vol.%Mo	1 785
$Pu_{0.5}Am_{0.5}O_{2-x}$ + 80 vol.%MgO	1 695
$Pu_{0.8}Am_{0.2}O_{2-x}$ + 75 vol.%MgO	1 535

HELIOS	
Composition	**T_{max} (K)**
$Am_2Zr_2O_7$ + 80 vol.%MgO	1 075
$Zr_{0.80}Y_{0.13}Am_{0.07}O_{2-x}$	995
$Pu_{0.04}Am_{0.07}Zr_{0.76}Y_{0.13}O_{2-x}$	1 845
$Am_{0.22}Zr_{0.67}Y_{0.11}O_{2-x}$ + 71 vol.%Mo	1 025
$Pu_{0.80}Am_{0.20}O_{2-x}$ + 84 vol.%Mo	1 610

Two fabrication processes that minimise contamination in the glove boxes were used to synthesise FUTURIX-FTA and HELIOS fissile phases: an oxalic co-precipitation route, and a combination of external gelation and infiltration methods [6]. The composite materials were then made with conventional powder metallurgy methods: mixing and grinding the non-radioactive powders with the fissile powders. The blends were sieved and pressed. The green pellets were then sintered. The two fabrication flow sheets for composite pellets are given on Figure 2; an illustration of the pellet microstructure is presented in Figure 3.

Figure 2: Schematic flow charts of oxide U-free fuels fabrication routes

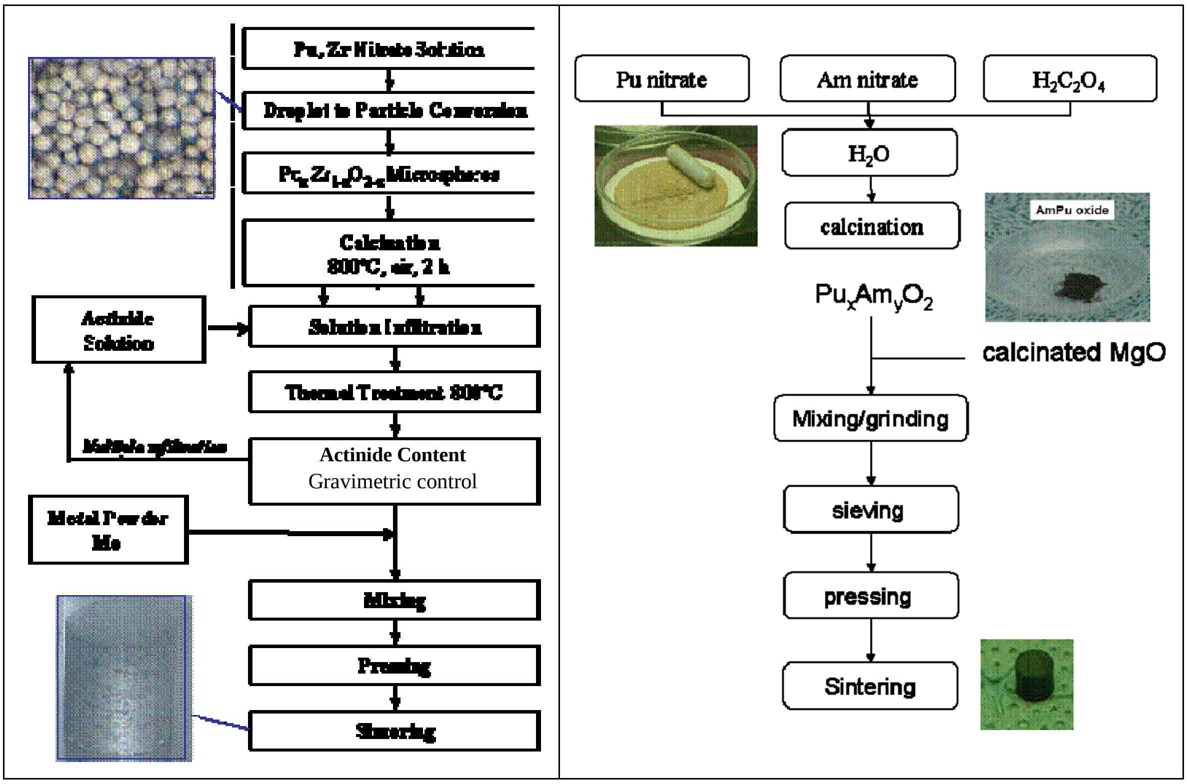
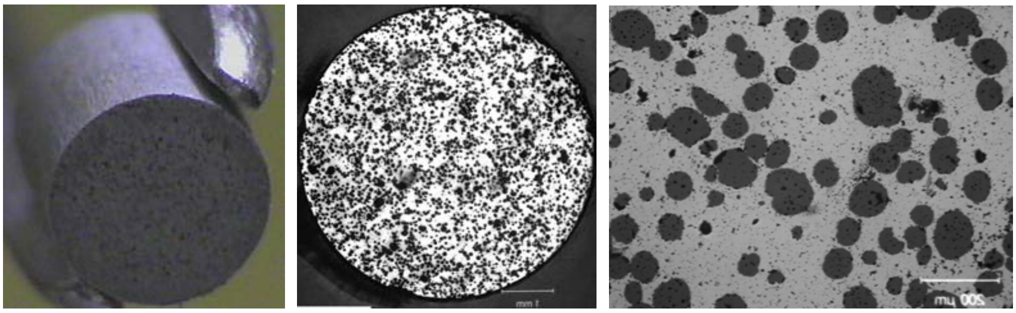


Figure 3: Visual aspect and microstructure of a CERMET pellet

($Pu_{0.23}Am_{0.24}Zr_{0.53}O_{2-x}$ + 60 vol.%Mo)



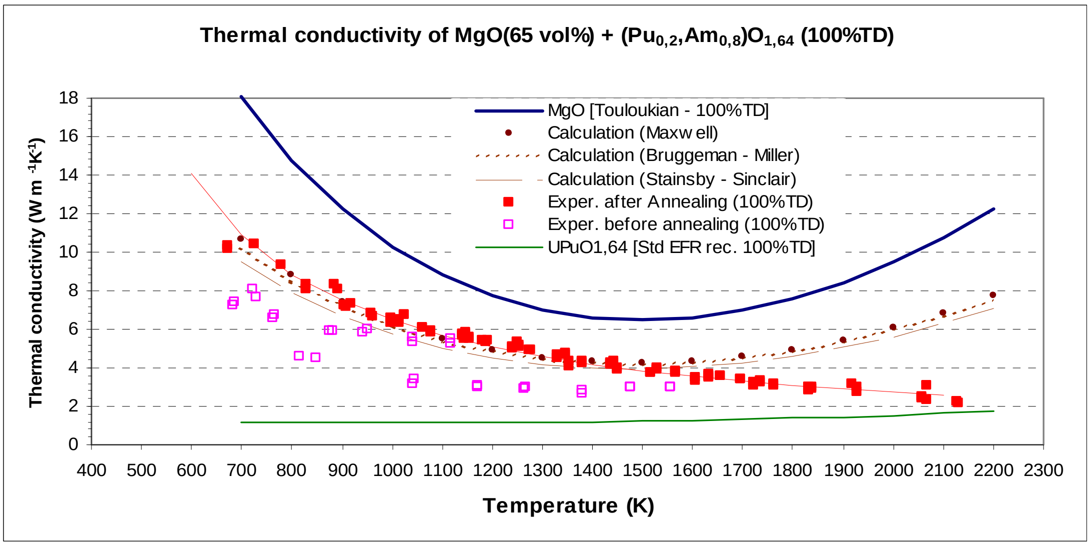
The BODEX experiment aims at investigating helium build-up and release mechanisms, with specific irradiation tests in HFR on boron-doped inert matrices stacked in instrumented pins in order to record the changes of pressure and temperature during the irradiation. Like ^{241}Am, ^{10}B produces under irradiation a large helium volume, but is easier to handle and to include in pins. After ~2 HFR cycles, 1.5 mmol of boron per cm^3 is sufficient for a representative helium production. Powders of borides (Mo_2B, ZrB_2) and borates ($Mg_3B_2O_6$) were mixed with respectively Mo, ZrO_2 and MgO powders. The blends were pressed and sintered. Blank pellets, ^{10}B- and ^{11}B-doped pellets were fabricated in order to discriminate the effects related to helium production from ^{10}B from those of irradiation (without boron) and chemical ones (^{11}B-doped compounds). The irradiation device was designed to reach both high and low temperatures (1 473 and 1 073 K) in the pellets. The irradiation is now completed. Post-irradiation examinations of the six irradiated capsules will start soon.

Out-of-pile measurements

Out-of-pile measurements aim at the acquisition of knowledge concerning the fuel thermal, mechanical and physico-chemical properties in order improve the databases with relevant and accurate data for the fuel design, fuel performance and safety modelling. Many results have been gained. Some specific points are given hereafter.

Thermal conductivity experimental measurements from 500 K to 2 200 K made on FUTURIX-FTA fuels have for example highlighted a significant drop at high temperatures (T > 1 500 K) for CERCER fuels, which were not predicted by calculations based on phase-mixing models (Figure 4).

Figure 4: Thermal conductivity of a CERCER fuel



Vaporisation measurements made on CERCER fuels in a Knudsen cell device have confirmed the low thermal stability of MgO under vacuum conditions (Figure 5). Volatilisation species and temperature ranges have been made clear too: the MgO vaporisation begins at 1 750-1 800 K, increases by five orders of magnitude between 1 800-1 900 K, and reaches a maximum at 2 400 K. Mg(g) is the major vaporised magnesium species, although MgO(g) has been detected within a magnitude of three decades lesser.

Chemical compatibility tests have been achieved [13] at 1 300 and 1 800 K under different atmospheres (air, Ar, Ar/H_2), between PuO_2 and AmO_2 and three inert matrices: MgO, Mo and Cr.[1] The results are as follows (see Table 6): under any atmosphere, no interaction is clearly observed between PuO_2 and MgO or Mo, whereas for both tests involving AmO_2 under argon, undetermined X-ray diffraction peaks are observed, revealing a different behaviour between Am and Pu. Tests with chromium all lead to the formation of perovskite type orthorhombic phases ($PuCrO_3$ and $AmCrO_3$) which definitively rule out Cr from the list of inert matrix candidates.

Conclusion

Studies on fuel development performed within the Integrated Project EUROTRANS, motivated by assessing the industrial practicability and transmutation of high-level nuclear wastes, have already provided a wide range of results.

Several cores have been designed and an optimum meeting the specification has been found. The thermo-mechanical calculations of the fuel pins loaded in the optimised cores have shown temperature and stress margins at beginning of life.

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[1] In order to confirm the chemical reactivity between Cr and actinide oxides, which has been suspected in the literature and which has led to discard Cr in a preliminary ranking.

Figure 5: Vaporisation of the components of the FUTURIX CERCER fuel

$Pu_{0.5}Am_{0.5}O_{2-x}$ + 80 vol.%MgO

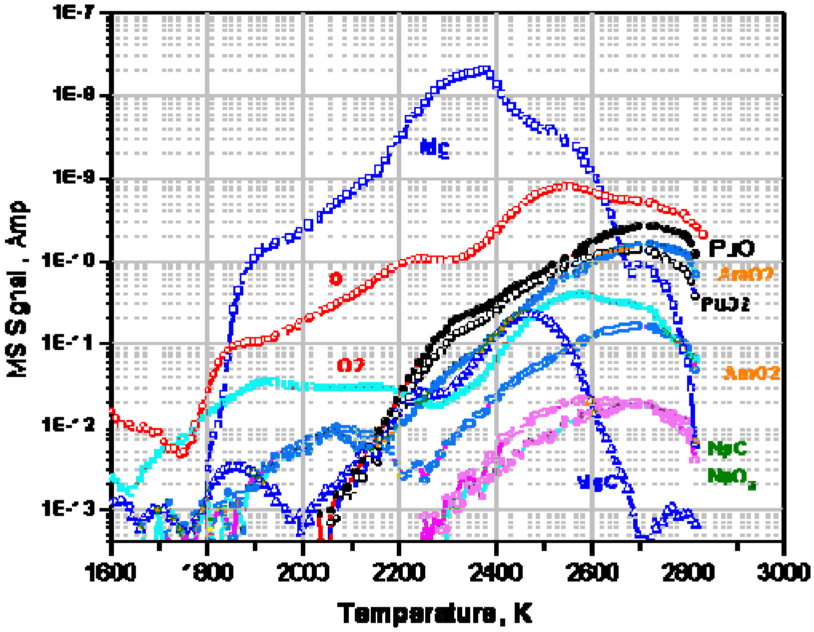

Table 6: Crystallographic identification of the products from the reaction between MgO, Mo and Cr matrices and fissile oxide phases at 1 300 K and 1 800 K

(h: hexagonal; c: cubic; m: monoclinic; o: orthorhombic)

Atm	PuO_2+MgO	AmO_2+MgO	PuO_2+Mo	AmO_2+Mo	PuO_2+Cr	AmO_2+Cr
Air	MgO PuO_2	AmO_2 MgO	–	–	–	–
Ar		Am_2O_3 h & c AmO_{2-x} c MgO Other peaks	PuO_2 Mo	Mo Am-Mo-O m?	PuO_2 $PuCrO_3$ o	AmO_{2-x} c $AmCrO_3$ o Cr Am_2O_3 h
Ar/H_2 5%	PuO_2 MgO	Am_2O_3 h & c MgO	PuO_{2-x} c Mo	Am_2O_3 h Mo		Cr_2O_3 $AmCrO_3$ o Am_2O_3 m & h

Transient calculations performed on the three-zone Mo-CERMET optimum core have pointed out that fuel and clad limits are not violated under conditions of ULOF and UTOP. For UBA simulations, pin failure would lead to a local voiding and reactivity addition but the fuel sweep-out effect leads to a power reduction and thus limits the core degradation.

The FUTURIX-FTA and BODEX irradiation tests are currently being performed in the PHENIX and HFR facilities. HELIOS pins should be loaded in HFR core before the end of this year.

Thermal properties of a large range of fuels have been experimentally evaluated: discrepancies between experiments and models previously used for the MgO-CERCER thermal conductivity description have been pointed out. Vaporisation measurements of the fuels in a Knudsen cell device have also been undertaken: the low temperature stability of MgO under vacuum conditions has been confirmed and the vaporisation mechanisms have been identified. Compatibility tests performed between fissile phases and inert matrix candidates have pointed out the reactivity of chromium, which makes it unsuitable as an inert matrix.

Up to the end of the project (March 2010), the programme will consist in:

- performing thermo-mechanical calculations during the whole residence time of the fuels in the cores;
- achieving the safety analyses for the optimised designs;
- achieving the irradiation tests and performing the post-irradiation examinations of BODEX and CONFIRM pins;
- performing fuel/clad and fuel/coolant compatibility tests, mechanical oxygen potential measurements on the fuels, and Pu-Am-O phase diagram investigations.

The most promising fuel will then be selected.

Acknowledgements

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Microstructural evolution and Am migration behaviour in Am-containing fuels at the initial stage of irradiation

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Abstract

In order to investigate the effect of americium addition to MOX fuels on the irradiation behaviour, the "Am-1" programme is being conducted in JAEA. The Am-1 programme consists of two short-term irradiation tests of 10-minute and 24-hour irradiations and a steady-state irradiation test. The short-term irradiation tests were successfully completed and the post-irradiation examinations (PIE) are in progress. The PIE for Am-containing MOX fuels focused on the microstructural evolution and redistribution behaviour of Am at the initial stage of irradiation and the results to date are reported. The successful development of fabrication technology with remote handling and the evaluation of thermo-chemical properties based on the out-of-pile experiments are described with an emphasis on the effects of Am addition on the MOX fuel properties.

Introduction

Uranium (U) and plutonium (Pu) mixed-oxide (MOX) fuels containing several per cents of minor actinides (MA) and fission products are promising candidates for a closed nuclear cycle system based on a fast reactor such as the JAEA Sodium-cooled Fast Reactor (JSFR) system which is part of the Fast Reactor Cycle Technology (FaCT) Development Project [1]. Although the content of MA in the MOX fuels varies mainly according to the required mass balance for MA, the maximum total content of MAs is considered to be 5% [2]. Americium (Am), which is representative of MA, should be considered important in view of its having a high generation yield in the spent fuel, a high and lasting radiotoxicity and expected impacts on various properties of MOX fuels. Am-containing MOX (Am-MOX) fuels, therefore, are being studied as a first step to promote research and development (R&D) of the MA-containing MOX (MA-MOX) fuels. R&D of Am-MOX fuels was started in the early 1990s in the Alpha-Gamma Facility (AGF) of the Japan Atomic Energy Agency (JAEA). Fabrication, characterisation and post-irradiation examinations of Am-MOX fuels have been successfully carried out for the purpose of obtaining fundamental insights into further development of MA-MOX fuels and many novel and practical results about Am effects on the fuel properties have been obtained at AGF.

This paper reports on the results of ceramography and electron probe microanalysis (EPMA) for Am-MOX fuel pellets at the initial stage of irradiation. It also presents the activities of the R&D programme for development of fabrication technology and out-of-pile properties for Am-MOX fuels used in the AGF.

Am-1 programme

In order to confirm the effect of MA addition on irradiation behaviour of MOX fuel pellets, an irradiation programme named “Am-1” is being conducted at the JAEA. The Am-1 programme consists of two short-term irradiation tests of 10-minute and 24-hour irradiations and a steady-state irradiation test. The objectives of the short-term irradiation tests are to confirm whether or not fuel melting occurs at a high linear heating rate and to evaluate the redistribution behaviour of Am during the initial burn-up. The steady-state irradiation test is expected to evaluate the behaviour of the fuel cladding chemical interaction (FCCI) and helium (He) release behaviour up to middle or high burn-up.

Two kinds of fuels were subjected to the irradiation tests of the Am-1 programme. One consisted of Am-MOX fuel pellets containing 3% or 5% Am and the other was MOX fuel pellets containing 2% Np and 2% Am (Np/Am-MOX). Am-MOX fuel pellets were fabricated using a remote handling technique in the shielded air-tight hot cell of the AGF at JAEA’s Oarai Research and Development Center. Np/Am-MOX fuel pellets were fabricated in a glove box unit of JAEA’s Plutonium Fuel Production Facility (PFPF) at Tokai Research and Development Center. Two values of the oxygen-to-metal ratio (O/M) of the fuel pellets were used as a test parameter in the Am-1 programme [3].

The two short-term irradiation tests are outlined as follows. Three Am-MOX fuel pins (which contained pellets for both Am percentages in different parts of the pins) and three Np/Am-MOX fuel pins were loaded into the B11(1) fuel subassembly by remote manipulation. This was done inside the hot cell of JAEA’s Fuel Monitoring Facility (FMF) at the Oarai Research and Development Center. The first irradiation test was conducted at a peak linear heating rate of about 43 kW/m for 10 minutes. Following the irradiation, one Am-MOX fuel pin and one Np/Am-MOX fuel pin were removed from the fuel assembly and were supplied for PIE. The remaining four fuel pins were reinstalled into another fuel subassembly, the B11(2), and then irradiated for 24 hours at almost the same peak linear heating rate as that of the first irradiation test. In the short-term irradiation tests, focus is being placed on elucidating the microstructural evolution and migration behaviour of Am in the MA-MOX fuels at the initial stage of irradiation.

The two short-term irradiation tests were successfully completed and the PIE have been conducted. A detailed description concerning the Am-1 programme, including fuel design and irradiation conditions, can be found in the literature [3,4].

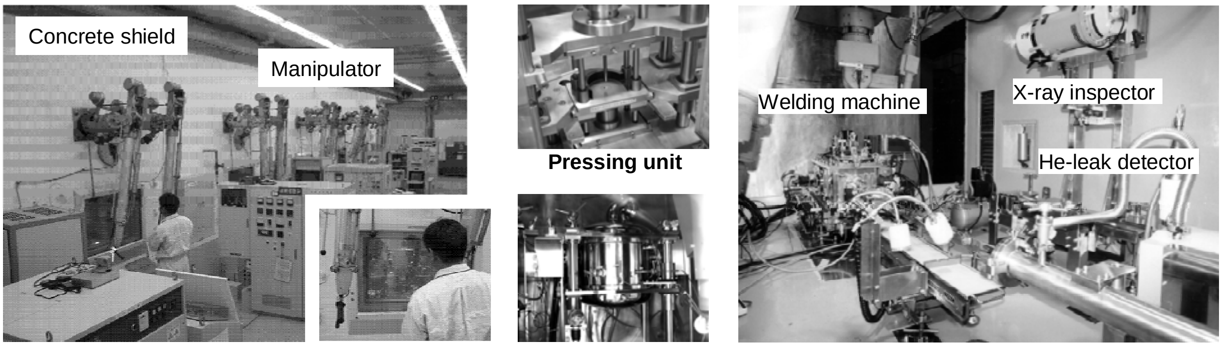
Fabrication technology and fundamental aspects of Am-MOX fuels

Fabrication technology with remote handling operation

It is difficult to handle Am in a glove box because it has strong gamma-ray radioactivity. Thus, the Am-MOX fuels must be fabricated in a shielded air-tight cell with remote handling in order to protect workers from gamma-ray exposure. This also means that control of maintenance and repair of the apparatuses must be done by remote operation. Figure 1 shows the operation area and some fabrication apparatuses installed in a shielded alpha-gamma type cell in the AGF. AGF was originally constructed as the PIE facility for irradiated fuels [5]. It was later equipped with a small-scale fuel fabrication unit in the hot cell for the purpose of developing remote fuel fabrication technology for MA-MOX fuels [6].

The apparatuses which were installed in AGF were systematically placed in the hot cell to ensure a smooth flow in the fabrication. The procedure is composed of powder treatment by rotation milling and mixing of raw powders, compaction by uni-axial pressing, and finally sintering in a reducing atmosphere. This fabrication procedure, which is conventional powder metallurgy, is relatively simple and compatible with remote operation. Materials used in the fabrication were transported by the conveyer system to make the operation easier. The apparatuses were remotely controlled from a panel in the operation room outside the hot cell as much as possible. Impurities in fabricated fuel pellets must be kept at a very low level since a small amount of impurities can degrade thermal properties of the fuel such as lowering the melting temperature and thermal conductivity. Therefore, inspection technology for the Am-MOX fuels was also established based on that of standard MOX fuel. The TIG welding machine, X-ray inspector and helium-leak detector were also installed for manufacturing of fuel pins and for confirmation of the integrity of the fuel pins. The handling by using manipulators made operators' exposure negligible and all apparatuses were designed so as to be easily maintained.

Figure 1: Operation area and fabrication apparatuses inside the hot shielded air-tight cell in the Alpha-Gamma Facility (AGF) of JAEA



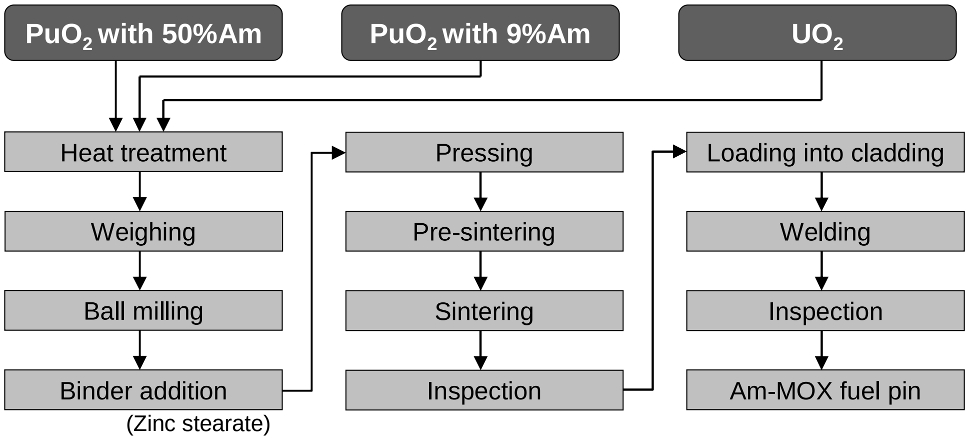
Operation area **Pressing unit** **Sintering Furnace** **Pin manufacturing and inspection**

Specifications and fabrication procedures of the Am-MOX fuels

Prior to the fabrication of Am-MOX fuels for the irradiation tests, many attempts were made to determine the effect of temperature patterns such as heating and cooling rates with controlled oxygen potential on the soundness of sintered pellets. As a consequence, optimal sintering atmospheres with controlled oxygen potential, together with suitable temperature control patterns, were established for the fabrication of Am-MOX fuel pellets. The annealing conditions for adjusting O/M ratio were also established.

Figure 2 shows a flow sheet of the fuel pin fabrication process. Powders of UO_2 and two kinds of Am-containing PuO_2 powders were used as raw materials. Detailed characterisation of the raw powders has been reported in the literature [6]. Before the sintering step, these three powders were annealed at 873 K for 2 h in a furnace with a Cantal heater to remove their absorbed moisture. After heat treatment, the powders were weighed by using an electronic balance in order to adjust the amount to the predetermined weight ratio. This was followed by mixing in a ball mill for 5 h. Zinc

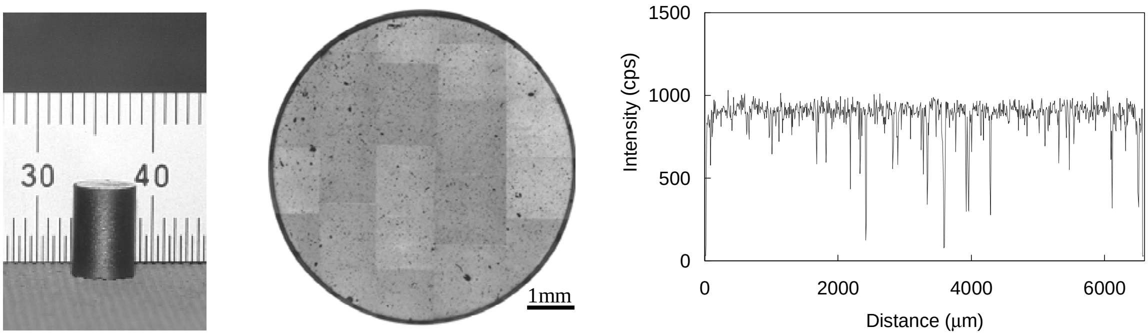
Figure 2: Fabrication flow of Am-MOX fuel pin



stearate was added as binder and the powder was further mixed for 30 min before cold-pressing on green pellets under a pressure of 3.84 t/cm^2. The green pellets were pre-sintered at 1 073 K for 2.5 h in the furnace with the Cantal heater to remove the binder and were sintered at 1 973 K for 5 h in a furnace with a tungsten mesh heater. The pellets were then heat treated to adjust the O/M ratio to the targeted values. The O/M ratio of the pellets was confirmed gravimetrically at room temperature. All of the heat treatments and sintering processes were carried out under the flowing gas atmosphere of Ar-5% H_2 or Ar-0.05% H_2 by adding an appropriate amount of moisture. After the sintering, fissile content, Pu enrichment, ^{235}U enrichment and Am content were determined using alpha spectrometry, gamma spectrometry and mass spectrometry. Volatile, moisture and metal impurities were also checked for the sintered pellets.

Figure 3 shows the visual appearance and a typical ceramograph of a 5% Am-MOX fuel pellet. Based on this image and others, Am-MOX fuel pellets which are crack-free and have a high density, homogeneous structure and good columnar configuration were obtained by adopting the optimal sintering condition. A homogeneous distribution of Am, as well as U and Pu, in the pellets was confirmed by EPMA. Crystal structure of Am-MOX fuels was revealed to be a single fluorite phase of the solid solution by X-ray diffraction (XRD) patterns.

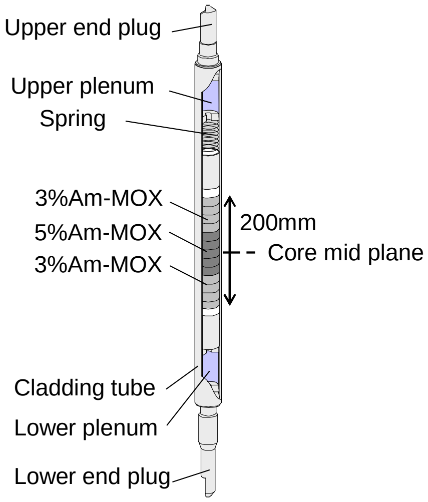
Figure 3: Outer appearance, ceramograph and Am distribution of a sintered 5% Am-MOX fuel pellet



Sintered pellet **Ceramograph** **EPMA line scanning (Am)**

The sintered and inspected Am-MOX fuel pellets were inserted into a cladding tube made of austenitic stainless steel, together with thermal insulator pellets of UO_2 and reflector components. The Am-MOX fuel pellets containing 5% Am were loaded at the middle of the fuel column and 3% Am-containing MOX fuel pellets were placed at the lower and upper ends of the fuel column. The fuel pins were sealed by TIG welding and then inspected for confirmation of their air-tightness/soundness. Figure 4 shows a schematic drawing and main specifications of the Am-MOX fuel pin. Manufactured fuel pins were then assembled into the fuel subassembly.

Figure 4: Configuration and main specifications of the Am-MOX fuel pin



Fuel pellet

Diameter: 6.52 mm
O/M ratio: 1.95 and 1.98
Density: 93% T.D.
Pu content: 30 wt.%
Am content: 3 and 5 wt.%

Cladding

Material: 15Cr-20Ni stainless steel
Outer diameter: 7.5 mm
Thickness: 0.4 mm

Fundamental studies

As well as the preparation of the fuel pellets for the irradiation test, fundamental studies have been carried out with emphasis on effects of Am addition on the thermo-physical/chemical properties of MOX in AGF. The phase relationship was investigated by XRD, differential thermal analysis (DTA), EPMA and ceramography on several Am-MOX samples having different Am contents and O/M ratios.

The lattice parameters of Am-MOX fuels were also determined from the XRD angles and were revealed to decrease with increasing Am content [7]. Melting point temperature was measured by a thermal analysis method [8]. Samples were taken from crushed pellets. The samples were loaded into tungsten capsules and were sealed under a vacuum of 1×10^{-2} Pa by electron beam welding. The capsule and sample were heated at a constant heating rate of about 20 K/min to about 3 100 K or higher with a high-frequency induction heating furnace. The measured data up to about 5% Am showed that the melting point temperature of Am-MOX fuels decreased with increasing Am content. The data were used in designing an irradiation test in Joyo [4]. The behaviour in the O/M ratio change of Am-MOX fuels during fabrication and storage in the air has also been investigated [9]. The weight of Am-MOX fuels was reported to increase relatively rapidly and its change rate strongly depends on the initial O/M ratio. The XRD pattern also changed with elapsed time after the annealing for O/M ratio control. That implied that the weight change of the fuel resulted from crystallographic change related to the O/M ratio. Oxygen potentials of Am-MOX were determined by thermogravimetry. It was reported that the addition of AmO_2, even in an amount as low as 4.5%, to MOX significantly increases the oxygen potentials [10].

Irradiation conditions

After the reactor power was raised continuously to the targeted peak linear heating rate of 43 kW/m and kept there for 10 min, the power was manually shut down. The fuel subassembly B11(1) was irradiated to around 0.0057% FIMA in the core centre location [000] of the operational cycle 3-1 of the Joyo MK-III core. The burn-ups for two samples of 3% Am-MOX and one sample of 5% Am-MOX were determined by chemical analysis based on the isotope dilution method [11] in the AGF. The fixed linear heating rate was evaluated using a fission rate derived from chemical analysis and the fission energy of each nuclide [12]. The determined linear heating rate agreed well with the calculated value [7] within an experimental error of 3%. The second irradiation test was performed at almost the same power history up to the targeted peak heating rate of 43 kW/m and held for 24 hours.

Experimental procedures of PIE

After the Joyo irradiation, the fuel subassemblies B11(1) and B11(2) were transported to the FMF adjacent to Joyo to carry out non-destructive PIE including X-ray CT scans [13], X-ray radiography, weight measurements, visual inspections, cladding outer diameter profilometry and gamma scanning. The results of non-destructive PIE showed that the Am-MOX fuel pins were irradiated without any failure as expected. After the non-destructive PIE, the fuel pins were punctured and sectioned to cut off the upper and lower ends of the fuel pins and then the pins were transported to the AGF for destructive PIE.

The Am-MOX fuel pin was cut at different axial positions into several segments. Several cross-sectional specimens were prepared by the following processes: each segment (about 20 mm in length) was impregnated with epoxy resin in vacuum and cut transversely into discs of about 5 mm in thickness. These disks were mounted into holders using epoxy resin, and then were ground and mirror-polished with anhydrous lubricant. The microstructure was observed with an optical microscope (TELATOM, Reichert).

EPMA of U, Pu and Am was done using the SX-100R model (Cameca) which was specially shielded with lead and tungsten to permit the analysis of irradiated nuclear fuels. Electron acceleration potential was 25 kV and beam current was 50 nA. Three wavelength dispersive spectrometers with pentaerythritol (PET) diffracting crystals were used for the analysis simultaneously. Characteristic X-rays selected were M-alpha lines for U and Am, and the M-beta line for Pu. The secondary electron image was used to obtain information about the fuel microstructure at the locations selected for analysis and to position the electron beam. Immediately prior to EPMA of the samples a thin film of carbon was applied to their surface by vacuum evaporation to avoid electron charging effects.

Results and discussion

Figures 5 and 6 show examples of as-polished macroscopic ceramographs of two Am-MOX fuel pellets irradiated for 10 min and 24 h. Some cracks were observed along the radial and circumferential directions on both specimens. Plano-convex-shape lenticular pores were observed around the central void of the specimen irradiated for 10 min. Lenticular pores were also found in the vicinity of cracks which developed in this fuel pellet due to thermal stresses. Crack healings occurred in some parts of both fuel pellets after the lenticular pores moved to the fuel centre along the temperature gradient. Closer inspection of the trails behind the lenticular pores (magnified views) showed that many of them consisted of a string of small spheres. The configuration of the lenticular pores was similar to that of conventional oxide fuels irradiated for a short time at the high linear heating rate condition [14-16]. No significant difference in the restructuring feature was observed between the 5% Am-containing and 3% Am-containing MOX fuel pellets. After the 24-h irradiation, the central void had developed to a size of about 1 mm in diameter. No lenticular pores were observed and highly dense columnar grains appeared on the specimen taken from around the axial mid position of the fuel pin. As shown in Figures 5 and 6, fuel restructuring had started in the Am-MOX fuel pellets even in the brief irradiation times of 10 min and 24 h.

Figure 5: Ceramographs of Am-MOX fuel irradiated for 10 min

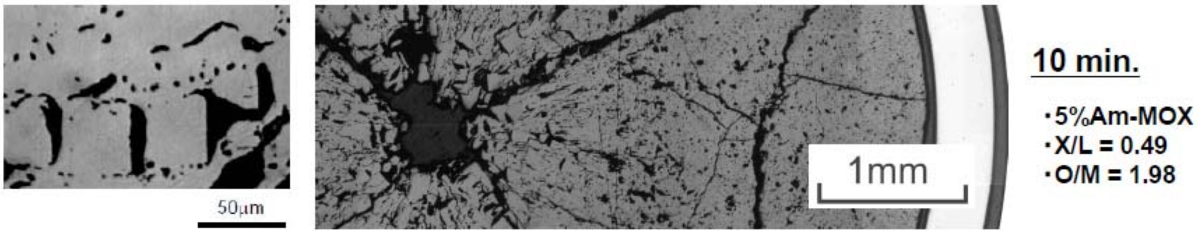
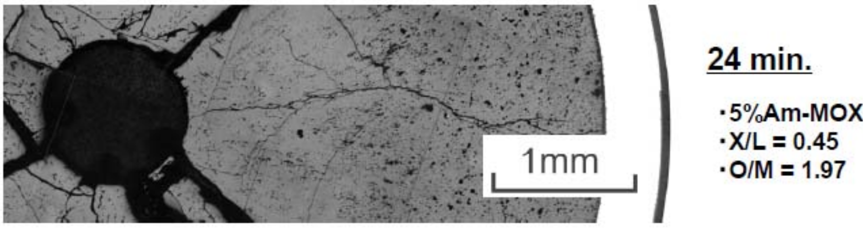


Figure 6: Ceramograph of Am-MOX fuel irradiated for 24 h



In order to confirm whether or not the fuel melting occurred at the high linear heating rate, careful observations were carried out. It has been reported that when the fuel melted at the initial stage of irradiation, a pore-free structure was observed in the centre of the fuel pellet or around the central void [17,18]. No sign of fuel melting was found in any of the specimens taken from the Am-MOX fuel pins irradiated for 10 min and 24 h. This implied that the thermal design for this test had a sufficient safety margin.

Figure 7 shows the radial distributions of U, Pu and Am for a specimen taken from the axial mid position of the fuel pin irradiated for 10 min. The concentration of these elements from the middle to outer part of the fuel pellets was almost the same as the chemically analysed value for an as-fabricated pellet. In the vicinity of the central void the concentrations of Pu and Am increased whereas that of U decreased. The radial distributions of U, Pu and Am on the specimens taken from the upper and lower ends of the fuel pin were quite flat and no apparent redistribution was found. The tendency for appearance or non-appearance of redistribution agreed with the fuel restructuring behaviour which could be explained by the axial distribution of linear heating rate. Figure 8 shows the radial distributions of U, Pu and Am on the specimen taken from the axial mid position of the fuel pin irradiated for 24 h. More obvious redistributions were measured on this specimen compared to the one irradiated for 10 min. The concentrations of Pu and Am gradually increased on approaching the edge of the central void. A slight increase in Am concentration at the centre of the fuel pellet has been reported in the Am-MOX fuel in the SUPERFACT programme [19].

Figure 7: EPMA results of U, Pu and Am distributions for 5% Am-MOX fuel specimen irradiated at about 43 kW/m for 10 min

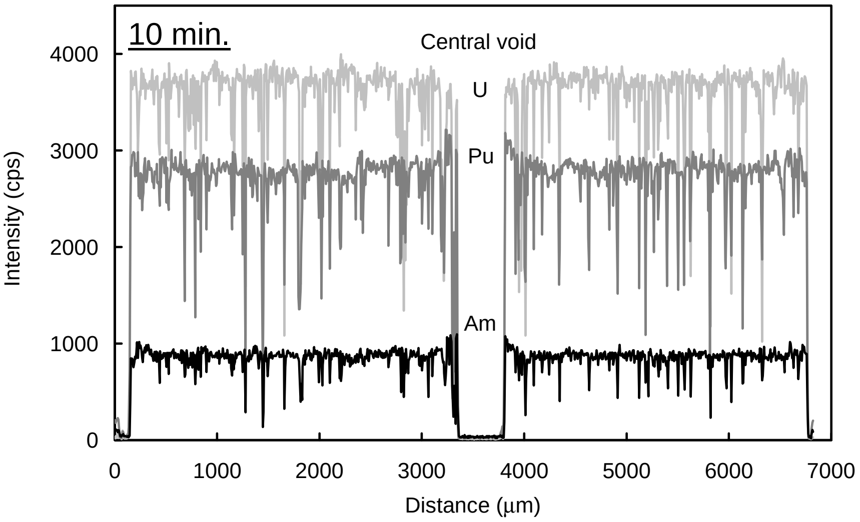
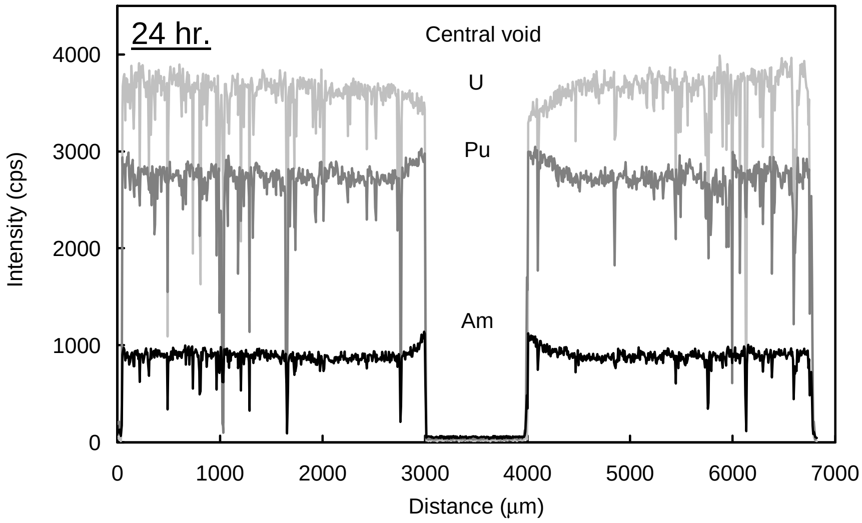


Figure 8: EPMA results of U, Pu and Am distributions for 5% Am-MOX fuel specimen irradiated at about 43 kW/m for 24 h



A quick comparison between Pu and Am distribution profiles shown in Figures 7 and 8 indicated both profiles were similar. Generally, ^{241}Am accumulates in Pu-containing fuels through the beta decay of ^{241}Pu. Therefore, careful attention should be paid to evaluating the Am concentration in MOX fuels. Calculation results obtained using the ORIGEN2 code confirmed the change in concentrations of Am and Pu was negligible during the short-term irradiation and the storage period up to the time of the EPMA analysis. That is to say, Am behaved like Pu during the initial stage of irradiation. It was considered that the increase in concentrations of Pu and Am in the centre of the fuel pellet could be attributed to vapour transport of U during the evaporation-condensation mechanism of the pores.

It is well known that the local concentration variations of Pu due to migration affect the fuel thermal properties in MOX fuel. It has been reported that the melting point temperature is decreased by the addition of a large amount of Am [8,20], which leads to a smaller design margin for fuel melting. The present EPMA results indicated that careful consideration must be given to the redistribution behaviour of Am as well as that of Pu for evaluating the impact on the thermal performance of Am-MOX fuels.

Summary

Remote fabrication technology of Am-MOX fuels was established in the AGF at JAEA. This technology is expected to be applied to the fabrication of future MA-MOX fuels. Fundamental studies have also been carried out emphasising the effects of Am addition on the thermo-physical/chemical properties of MOX.

Short-term irradiation tests of Am-MOX fuels were conducted. The results of non-destructive PIE showed that all of the Am-MOX fuel pins were irradiated without any failure as expected. The microstructural evolutions were observed by optical microscopy and redistribution behaviour of constituent elements were determined by EPMA. The ceramography results showed that structural changes such as formation of lenticular voids and the central void occurred quickly and early within a 10-minute irradiation. The results of EPMA revealed that Am migrated to the radial centre of the fuel pellet along the temperature gradient during the initial stage of irradiation. This tendency is similar to that of Pu. The irradiation behaviour at the high linear heating rate obtained in the Am-1 programme will be useful for the modelling and design study of Am-containing MOX fuels.

The technologies and data as well as experiences and findings obtained through the R&D programme for developing Am-MOX fuels can be reflected onto the development of the MA-containing MOX fuels for the closed nuclear cycle based on fast reactor use.

Acknowledgements

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Development of nitride fuel and pyrochemical process for transmutation of minor actinides

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Abstract

Nitride fuel cycle for transmutation of minor actinides has been investigated under the double-strata fuel cycle concept. Mononitride solid solutions containing minor actinides have been prepared and characterised. Thermo-physical properties, such as thermal expansion, heat capacity and thermal diffusivity, have been measured by use of minor actinide nitride and burn-up simulated nitride samples. Irradiation behaviour of nitride fuel has been examined by irradiation tests. Pyrochemical process for treatment of spent nitride fuel has been investigated mainly by electrochemical measurements and nitride formation behaviour in pyrochemical process has been studied for recycled fuel fabrication. Recent results of experimental study on nitride fuel and pyrochemical process are summarised in the paper.

Introduction

Nitride fuel has been proposed as a candidate fuel for the accelerator-driven system (ADS), since it has superior thermal and neutronic properties suitable for transmutation of minor actinides (MA) [1]. Formation of the solid solution over a wide range of actinide combinations and compositions is another important advantage of nitride fuel for transmutation. On the other hand, the spent nitride fuel of ADS is to be reprocessed using a pyrochemical process for recycling MA and Pu, since it has several advantages in case of treating spent fuel with large decay heat and fast neutron emission. The Japan Atomic Energy Agency (JAEA) has undertaken fundamental R&D on nitride fuel and the pyrochemical process in the framework of the double-strata fuel cycle concept.

MA-bearing nitrides have been prepared by carbothermic reduction of the dioxide. Sintered pellets for thermo-physical property measurements and irradiation tests have been prepared by a classical powder metallurgical manner. In case of a so-called U-free nitride fuel for ADS, MA is contained as a principal component and a diluent material is added to the fuel in place of U. For the moment, ZrN has been chosen as the first candidate of the diluent material and TiN as the second one in JAEA. Mononitride solid solutions containing not only MA but also ZrN have been prepared and characterised. It was confirmed that the addition of ZrN contributes to the improvement of chemical stability of nitride fuel with hygroscopic nature. Further, U-based burn-up simulated nitride pellets were prepared and subjected to thermo-physical property measurements in Osaka University besides in JAEA. On the other hand, R&D activities on pyrochemical process include anodic dissolution of nitride fuel in LiCl-KCl molten salt, recovery of MA and Pu in liquid Cd cathode and re-nitridation of actinides recovered in liquid Cd cathode. Besides MA and Pu nitrides, electrochemical behaviours of U-based burn-up simulated nitrides and ZrN or TiN-containing nitrides have been examined. The nitridation-distillation combined reaction for re-nitridation of actinides was developed and applied to the formation of PuN, AmN and (U,Pu)N.

Under an extensive experimental study by use of MA and Pu, technical feasibility of nitride fuel cycle for transmutation of MA has been demonstrated in a laboratory scale. This paper summarises recent results of experimental study on nitride fuel and pyrochemical process, mainly focusing on those obtained after the last conference in Nîmes [2].

Preparation of MA nitrides

Following the preparation of MA mononitrides and their binary or ternary solid solutions, a quaternary solid solution of (Np,Pu,Am,Cm)N was prepared by carbothermic reduction of the mixture of $^{237}NpO_2$, $^{243}AmO_2$ and $(^{240}Pu,^{244}Cm)O_2$. In this case, a starting C/MO_2 (M=Np, Pu, Am, Cm) molar ratio was adjusted at 3.91 and the carbothermic reduction was carried out at 1 573 K for 4.8 ks in N_2 stream, followed by the removal of residual carbon and oxygen in N_2-H_2 mixed gas stream at 1 773 K for 9 ks and at 1 793 K for 25.2 ks. The product of carbothermic reduction was subjected to chemical and X-ray diffraction analyses.

Figure 1 shows the X-ray diffraction pattern of (Np,Pu,Am,Cm)N and the results of chemical and X-ray diffraction analyses are summarised in Table 1. The formation of quaternary mononitride solid solution was demonstrated for the first time, as shown in Figure 1. Further, the lattice parameter of 0.4943 nm almost agreed with the value assumed from the Vegard's law between NpN-PuN-AmN-CmN. Impurity contents of oxygen and carbon could be kept at 0.11 wt.% and less than 0.04 wt.%, respectively.

The preliminary results of a sintering test for (Np,Pu,Am,Cm)N suggested that dense pellets higher than 85% TD could be obtained without significant loss of Am by sintering green pellets in a N_2-H_2 mixed gas stream at a temperature lower than 1 953 K. The appearance of the sintered disk of (Np,Pu,Am,Cm)N with 86% TD is shown in Figure 2.

The dedicated fuel for transmutation of MA is a so-called U-free fuel and a diluent material is added in place of U. As for the diluent material, JAEA has proposed ZrN as the first candidate for ADS and TiN as the second one. Both ZrN and TiN have a higher thermal conductivity and melting temperature than actinide mononitrides and a stable structure without phase transformation. These characteristics are favourable for design of a dedicated fuel for ADS, in addition to other general requirements such as neutronic property and resistance against radiation damage summarised elsewhere [3]. Typical characteristics of ZrN and TiN are shown in Table 2 in comparison with PuN.

Figure 1: X-ray diffraction pattern of (Np,Pu,Am,Cm)N prepared by carbothermic reduction

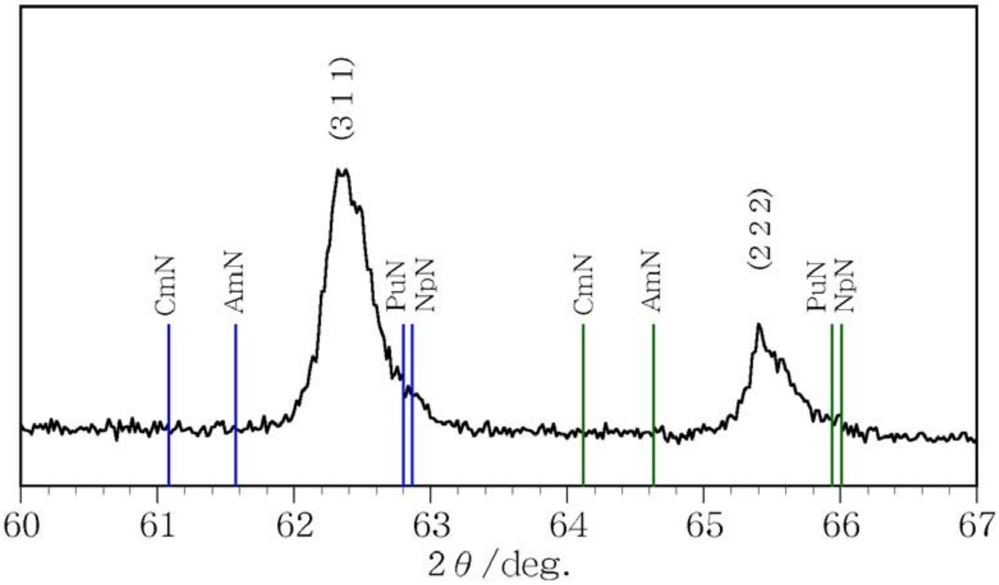


Table 1: Results of chemical and X-ray diffraction analyses for (Np,Pu,Am,Cm)N prepared

Nominal composition	Chemical analysis			X-ray diffraction analysis	
	N content (wt.%)	O content (wt.%)	C content (wt.%)	Phases identified (nm)	Lattice parameter
$(Np_{0.279}Pu_{0.307}Am_{0.279}Cm_{0.135})N$	5.87	0.11	< 0.04	Mononitride	0.4943

Figure 2: Appearance of (Np,Pu,Am,Cm)N disk with 86% TD

Table 2: Typical characteristics of ZrN and TiN in comparison with PuN

	ZrN	TiN	PuN
Thermal conductivity (W/m/K) 1 273 K 1 773 K	 ~23 ~26	 ~60 ~65	 ~11 ~12
Melting temperature (K)	3 253	3 205	2 843
Crystal structure	NaCl-type fcc	NaCl-type fcc	NaCl-type fcc
Lattice parameter (nm)	0.4578	0.4242	0.4905

Although ZrN and TiN have the same crystal structure, ZrN is likely to form a solid solution with actinide mononitrides, while the mutual solubility of TiN and actinide mononitrides is negligibly small. So the nitride fuel with ZrN will be a solid-solution type fuel, while the nitride fuel with TiN will be a composite type fuel. These characteristics are derived from the difference of their lattice parameters and those of actinide mononitrides [4]. Although it was found that 0.3(Pu,Am,Cm)N-0.7ZrN sample with high carbon impurity content had two *fcc* phases with different (Pu+Am+Cm)/Zr ratios [5], the ADS fuel designed by JAEA, whose composition is proposed as $(MA_{0.24}Pu_{0.16}Zr_{0.60})N$, would be a single-phase solid solution. It was found that the addition of ZrN contributes to the increase of chemical stability of MA nitride with hygroscopic nature at room and elevated temperatures [6]. The preparation of MA-bearing nitride pellets containing ZrN or TiN is underway.

Properties of MA nitrides and burn-up simulated nitrides

Thermo-physical properties of MA-bearing nitrides, which are necessary for designing the ADS fuel and understanding the irradiation behavior, had not been available. So the thermal expansions, heat capacities and thermal diffusivities were measured by high-temperature X-ray diffractometry, drop calorimetry and laser flash method, respectively. The thermal conductivities were obtained from the product of thermal diffusivity, heat capacity and bulk density.

Lattice parameters of NpN, PuN and AmN were measured from room temperature to 1 478 K by high-temperature X-ray diffractometry. It was found that the linear thermal expansions of AmN and PuN were close to each other and larger than that of UN, while that of NpN was nearly the same as UN [7]. Measurements on (Np,Am)N, (Pu,Am)N, (Np,Pu,Am,Cm)N and (Pu,Am,Zr)N were also carried out. The results suggested that the average thermal expansion coefficient of the solid solution could be approximated from that of the constituent nitrides by use of the linear mixture rule [8].

The enthalpy increments of NpN and AmN were measured with a twin-type drop calorimeter from 354 to 1 071 K, as shown in Figure 3. Heat capacities were determined by the derivatives of the quadratic polynomial equation for the enthalpy increment with respect to temperature. Figure 4 shows the heat capacities of NpN and AmN in comparison with literature values for UN and PuN [9]. It was seen that heat capacity of NpN was nearly the same as those of UN and PuN, while that of AmN was slightly smaller than those of UN, NpN and PuN. Heat capacities of actinide mononitrides had similar temperature dependence. The measurements for the solid solutions containing MA are also planned for testing the validity of the Neumann-Kopp rule.

Figure 3: Enthalpy increment values of NpN and AmN

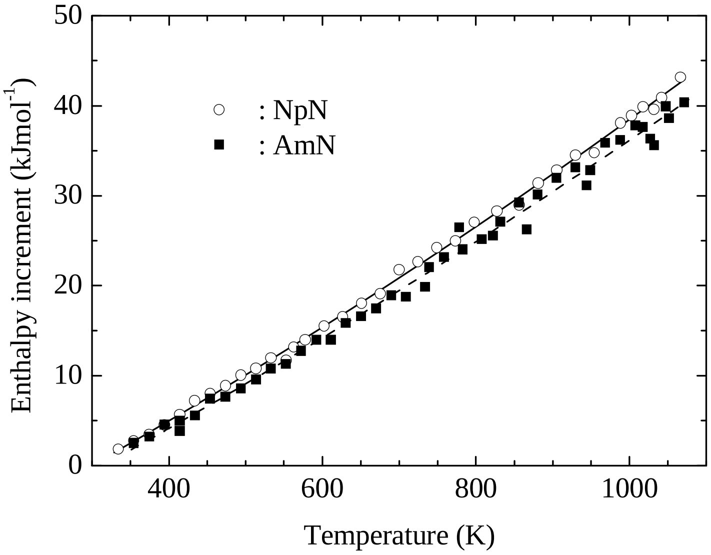
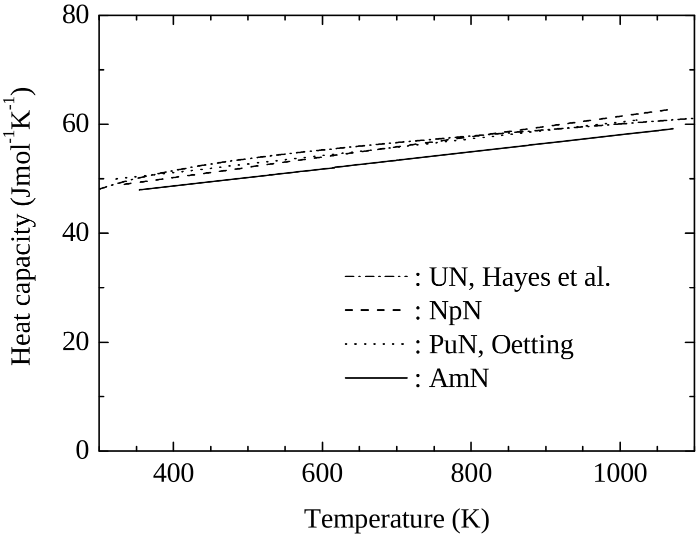


Figure 4: Heat capacities of NpN and AmN together with those of UN and PuN



The thermal diffusivities of NpN and AmN were measured from 373 to 1 473 K, from which the thermal conductivities were calculated. The thermal conductivity of NpN corrected to 100%TD almost agreed with the literature value [10] and that of AmN was obtained for the first time [11]. The thermal conductivities of UN, NpN, PuN and AmN had similar temperature dependence that they gradually increase with temperature over the temperature range investigated. It was also found that the thermal conductivities decreased with atomic number of actinides from UN to AmN. The measurements for the solid solutions not only MA but also ZrN are underway for clarifying the composition dependence. Preliminary results suggested that the addition of ZrN to the solid solution increases the thermal conductivity especially at elevated temperatures.

Other R&D activities include measurements of thermo-physical properties of U-based burn-up simulated nitrides. Non-radioactive fission product elements of (Nd, Mo) as well as possible diluent materials (ZrN, TiN) were added to uranium nitride so as to examine the effects of the fission products and diluent materials on the properties. The samples were characterised by X-ray diffractometry and scanning electron microscopy/energy dispersive X-ray analysis. The heat capacity was measured by triple cell type of differential scanning calorimeter, and the thermal diffusivity was measured by the laser flash method. The thermal conductivity derived from the thermal diffusivity and heat capacity was found to decrease with the addition of NdN, while the addition of Mo did not apparently affect the thermal conductivity. The thermal conductivities of (U,Nd)N, UN+Mo and (U,Nd)N+Mo pellets are displayed in Figure 5 [12]. Also in the case of (U,Nd)N+Mo, the thermal conductivity was apparently affected only by the addition of Nd. Mechanical property of thermal creep for the burn-up simulated nitrides was measured by the compressive test. The steady-state creep rate of UN was found to decrease with the addition of NdN, and a similar result was obtained for (U,Zr)N [13]. Young's modulus and Vickers hardness were also measured, from which the dependence of Young's modulus on the porosity and the dependence of hardness on the load were clarified [14].

Irradiation behaviour of nitride fuel

Following the irradiation test of (U,Pu)N fuel pins in JMTR and fast test reactor JOYO, the irradiation test of U-free nitride pin was carried out at JAEA. (Pu,Zr)N and PuN+TiN pellets were fabricated and the fuel pin containing the pellets was irradiated in JMTR in order to examine the effect of the diluent materials on the irradiation behaviour. The structure of fuels pin and the irradiation condition are shown in Figure 6 and Table 3, respectively. Post-irradiation examinations (PIE) have been completed in JAEA's hot cells. For the moment, no detrimental effects of the addition of diluent materials have been found from the results of the PIE [15].

Figure 5: Thermal conductivities of U-based burn-up simulated nitrides containing Nd and Mo

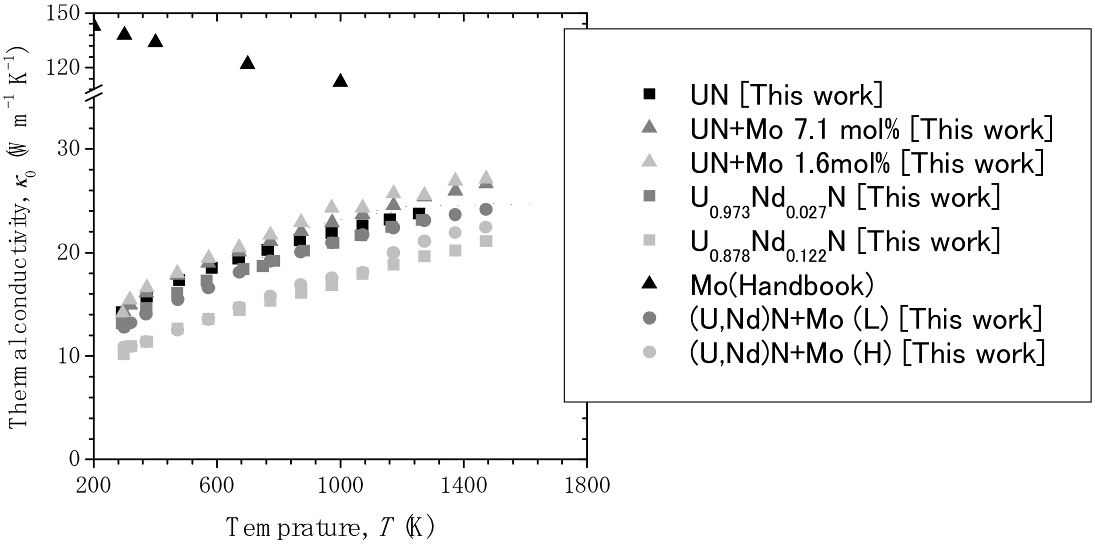


Figure 6: Structure of U-free nitride fuel pin containing (Pu,Zr)N and PuN+TiN pellets

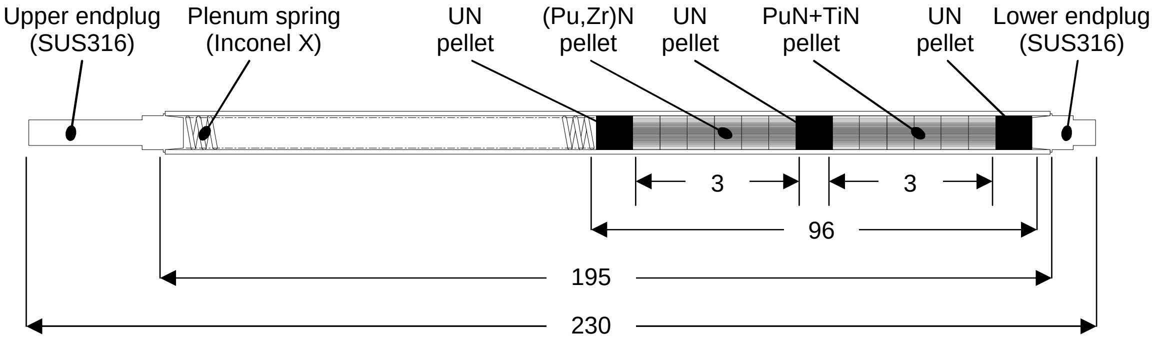


Table 3: Irradiation condition of U-free nitride fuel pin containing (Pu,Zr)N and PuN+TiN pellets

	(Pu,Zr)N	PuN+TiN
Pu content	20 wt.%	20 wt.%
Linear power	408 W/cm	355 W/cm
Peak burn-up	14.7 at.%(Pu)	17.0 at.%(Pu)
Max. fuel temperature (estimation)	1 273 K	1 083 K

Among the results of PIE, the radial distributions of Pu after irradiation are shown in Figures 7 and 8 for (Pu,Zr)N and PuN+TiN pellets, which reflected the characteristics of a solid-solution type (Pu,Zr)N fuel and a composite type PuN+TiN fuel.

The irradiation test of transmutation fuel with high MA content has been in progress under the international collaboration programme FUTURIX-FTA [16]. In this programme CERCER and CERMET type oxides, metal and nitride fuels are under irradiation in Phénix. JAEA is involved in the nitride fuel programme of FUTURIX-FTA. This programme is the first irradiation test of MA-bearing nitride fuel under fast neutron circumstances, so valuable information on the irradiation behaviour is expected from the results of the PIE.

Figure 7: Radial distribution of Pu in (Pu+,Zr)N pellet after irradiation

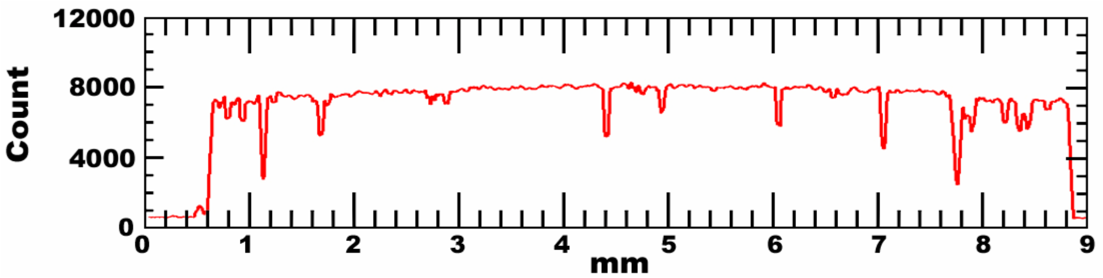
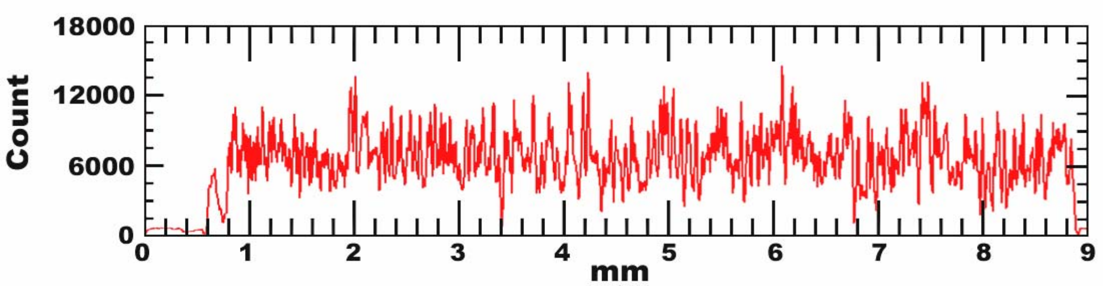


Figure 8: Radial distribution of Pu in PuN+TiN pellet after irradiation



Pyrochemical process

Spent nitride fuel for ADS is treated by a pyrochemical process for recycling MA and Pu. Anodic dissolution behaviour of actinide mononitrides in LiCl-KCl molten salt was investigated using electrochemical measurements, where the evolution of nitrogen as N_2 gas was confirmed by gas chromatography [17]. The equilibrium potentials were measured and analysed by the thermodynamic relationship assuming a partial nitrogen pressure in the molten salt. It was found that the equilibrium potentials of AnN (An=U, Np, Pu, Am) were positively shifted in comparison with the redox potentials of An^{3+}/An couple and these potential shifts almost corresponded to the free energy of formation of their mononitrides. Further, it was demonstrated that actinides were recovered in solid or liquid Cd cathode by the potential controlled electrolysis of their mononitrides.

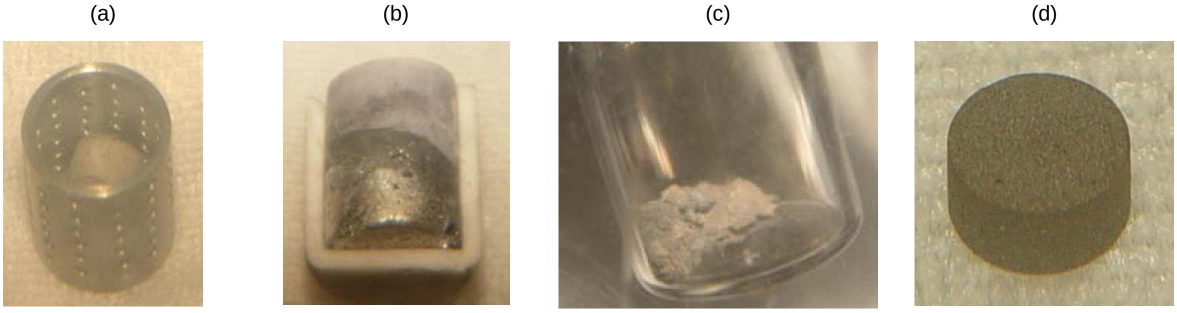
MA and Pu recovered in liquid Cd cathode are to be converted to nitride again in the nitride fuel cycle for ADS. It was observed that actinides beyond their solubility limit in liquid Cd formed intermetallic compounds with Cd. The nitridation-distillation combined reaction was developed for the Pu-Cd, Am-Cd and U-Pu-Cd alloys, where the nitridation of actinides and the distillation of Cd proceeded simultaneously by heating the alloy in N_2 gas stream at 973 K [18]. Further, the pellet preparation was carried out by use of the nitride powder formed by the nitridation-distillation combined reaction.

A series of experiments, the potential controlled electrolysis of (U,Pu)N with liquid Cd cathode, the nitridation-distillation combined reaction of U-Pu-Cd alloy in liquid Cd cathode, milling the powder product of the reaction, pressing into green pellet and sintering, were carried out. Figure 9 shows the appearance of the (U,Pu)N pellet before electrolysis, liquid Cd cathode after electrolysis, (U,Pu)N powder formed by the nitridation-distillation combined reaction and sintered (U,Pu)N pellet. According to preliminary characterisation results, the sintered pellet had an almost single phase of (U,Pu)N and density of about 84% TD. The oxygen impurity contents ranged from 0.1 to 0.2 wt.% [19].

The other R&D activities of concerning the pyrochemical process deal with the anodic dissolution behaviour of U-based burn-up simulated nitride and ZrN or TiN-containing nitride samples. UN pellets containing Mo, Nd or Pd were subjected to electrochemical measurements in LiCl-KCl-UCl_3 melt and the potential controlled electrolysis by use of liquid Cd cathode. It was found that most of the U in the burn-up simulated nitrides was dissolved in the salt at a similar anode potential applied

Figure 9: Appearances of the sample in a series of experiments

(a) (U,Pu) pellet before electrolysis, (b) liquid Cd cathode after electrolysis, (c) powder formed by the nitridation-distillation combined reaction, (d) sintered (U,Pu)N pellet prepared from the recovered powder



to the electrolysis of UN and recovered in liquid Cd cathode. On the other hand, Mo and Pd in the sample remained in anode undissolved, while Nd was dissolved but most of them stayed in the salt phase un-recovered [20]. (U,Zr)N and UN+TiN pellets were also subjected to electrochemical measurements in LiCl-KCl-UCl_3 melt. It was found that U in UN+TiN pellet was dissolved in the salt at the same anode potential applied to the electrolysis of UN, while a more positive potential was necessary for the dissolution of U in (U,Zr)N. Further, the equilibrium potential in LiCl-KCl-UCl_3 melt depended on the ZrN content in (U,Zr)N [21]. The results reflected the thermodynamic stabilisation of (U,Zr)N due to the formation of solid solution.

Conclusion

In order to develop nitride fuel cycle for transmutation of long-lived MA, fundamental R&D on the preparation of MA nitride fuel, the thermo-physical property measurements, the irradiation test of nitride fuel and the development of a pyrochemical process have been carried out. As described in the paper, R&D has progressed through the experimental study of not only MA and Pu but also burn-up simulated samples, from which the feasibility of a nitride fuel cycle for the transmutation of MA was demonstrated at laboratory scale.

Acknowledgements

The present study includes the results of the study entitled *Technological Development of a Nuclear Fuel Cycle Based on Nitride Fuel Cycle and Pyrochemical Reprocessing* entrusted to JAEA by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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The RIAR DOVITA-2 P&T programme – results of 15 years of R&D activity*

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Abstract

In 2007, RIAR modified the well-known demonstration DOVITA programme to reflect a new generation research and development P&T programme concept designated DOVITA-2 (dry technologies for MA fuel reprocessing, on-site disposition of SNF reprocessing and fabrication of various types of fuels and fuel pins, integration of MA recycling option into close fuel cycle of industrial fast reactors, the whole complex of approaches will permit a creation of the compact plant for transmutation of actinides). The DOVITA-2 programme should demonstrate opportunities of different technologies for realisation of an optimised fuel cycle for the actinide burner fast reactor (ABFR) (for transmutation of Np, Am, Cm).

RIAR uses the BOR-60 reactor as an experimental base for DOVITA-2 investigations, as the hard BOR-60 neutron spectrum is very beneficial for minor actinide burning.

The DOVITA-2 programme is currently under way. The investigations carried out over the past 15 years have shown that most of the problems connected with the ABFR fuel cycle can be solved. The problem of minor actinide recycle can be solved without development of new exotic systems, and only on the basis of the known technological methods and with the application of well-known reactor systems.

The paper contains the data on the complex study of the chemical aspects of the programme and the demonstration reactor experiments which will be completed in the coming years.

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* The full text of this paper was unavailable at the time of publication.

Experimental evaluation of Am- and Np-bearing mixed-oxide fuel properties

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Abstract

Homogeneous MOX fuels containing minor actinide elements such as Np and Am have been developed for the advanced Japan sodium-cooled fast reactor. In this report, the physical properties, i.e. melting temperature, thermal conductivity, lattice parameter, oxygen potential and phase separation behaviour, of the fuel were measured and effects of MA content were discussed. Melting temperatures of Am-bearing MOX were measured and the Am addition was estimated to decrease melting temperature by 4 K per 1% Am content. The effect of Np content was evaluated by using a newly expanded ideal solution model of the UO_2-PuO_2-AmO_2-NpO_2 *system. Thermal conductivities of Am and Np-bearing MOX were evaluated as functions of densities, MA contents, oxygen-to-metal ratios and temperatures. Thermal conductivities decreased significantly with decreasing density and oxygen-to-metal ratio. The Am and Np contents were observed to lower thermal conductivities slightly at temperatures lower than 1 000 K. The lattice parameters of* $(U,Pu,Am,Np)O_{2.00}$ *were measured, and the model was derived. The change of the theoretical density through the addition of MA was negligible. Oxygen potentials of* $(U_{0.8}Pu_{0.2})O_{2-x}$, $(U_{0.7}Pu_{0.3})O_{2-x}$ *and* $(U_{0.66}Pu_{0.3}Am_{0.02}Np_{0.02})O_{2-x}$ *were measured previously at temperatures of 1 473-1 623 K. Those of* $(U_{0.66}Pu_{0.3}Am_{0.02}Np_{0.02})O_{2-x}$ *were slightly higher than those of MOX without MA. Fuel-cladding chemical interaction was not significantly affected by adding the minor actinides. The effect of phase separation of hypo-stoichiometric MOX was evaluated. The temperature of phase separation decreased with increasing Am and Np contents and the effect of Am content was larger than that of Np content. Phase separation occurred at temperatures below the coolant temperature, and no phase separation affected the irradiation behaviour. In conclusion, additions of Am and Np did not significantly affect fuel properties.*

Introduction

It is essential in developing advanced nuclear reactor technology to discuss how to deal with the minor actinides (MA) produced in the fuels. Various MA handling methods have been proposed for development. Japan Atomic Energy Agency (JAEA) has proposed the nuclear recycle system in which homogenous MOX fuels containing MA have been used in the Japan sodium-cooled fast reactor (JSFR) [1-4]. Their fuel design is aimed at raising nuclear proliferation resistance and reducing effects on the environment by extracting and recycling MA from spent fuel.

In the development of such a new type fuel, it is necessary to measure fuel physical properties as a parameter of composition for the fuel design and the irradiation behaviour evaluation. The MOX fuels contain up to 5% MA content, mainly Np and Am, and a small amount of Cm of less than 0.5%. In addition, the oxygen-to-metal (O/M) ratio of the fuels which is an important parameter to control the fuel and cladding chemical interaction is adjusted to about 1.95 to attain high burn-up. The temperature of the fuels is over 2 300 K because of the high linear heating rate. The large temperature gradient causes redistribution of MA and the O/M ratio in the radial direction in pellets. Therefore measuring the physical properties and evaluating the effects of MA content and O/M ratio are necessary.

Many studies on the physical properties of fast reactor MOX fuel were carried out in the 60s to 80s, but the data are scattered (see for example [5-7]). Recently, physical properties of MA-bearing MOX have been reported. These studies include accurate measurements of melting temperatures [8-12], thermal conductivities [13-17], lattice parameters [18,19], oxygen potentials [20-22] and phase separation temperature [18,19]. As well, the effects of MA contents were evaluated. These recent measurement results are summarised here and the effects of Np and Am contents on fuel properties are discussed using a newly expanded model.

Melting temperatures

An early study [5] related to the melting process of oxide fuels was carried out by the V-filament method. During measurement, a sample composition change was caused by loss of gaseous species of U and Pu, and consequently the data had large error. Shortly after, the thermal arrest method was developed, which made measurements on a using sample encapsulated in tungsten (W). Two groups measured the melting temperatures of MOX systematically by that method [6,7]. Unfortunately, analyses of the melted samples were not carried out in their measurements.

Recently, Kato, *et al.* [8,9] found that a reaction occurred between a sample and W capsule during measurements by the thermal arrest method, and the measured high Pu content MOX were not true. They used a rhenium (Re) inner capsule to prevent sample reaction with the W capsule and measured the melting temperatures as functions of Pu and Am contents [8].

The phase diagram of the UO_2-PuO_2 system has also been revised. New measurement results showed that the solidus temperature of MOX with 40% Pu was at 3 001 K ± 35 K [8] which was 56-93 K higher than other earlier works [6,7]. The ideal solid solution model was the basis for a new model to calculate the melting temperatures in the UO_2-PuO_2-AmO_2 system [8]. This derived model reproduced the experimental solidus temperatures with an accuracy of ±20 K. Now, the authors have expanded the model to reproduce the melting temperature in the UO_2-PuO_2-AmO_2-NpO_2 system.

In the quarternary solid solution, compositions and melting temperatures are obtained by simultaneously solving the following equations:

$$\frac{\Delta h_m(UO_2)}{R}\left(\frac{1}{T_m(UO_2)}-\frac{1}{T_m}\right)=\ln\left(\frac{x^l(UO_2)}{x^s(UO_2)}\right) \tag{1}$$

$$\frac{\Delta h_m(PuO_2)}{R}\left(\frac{1}{T_m(PuO_2)}-\frac{1}{T_m}\right)=\ln\left(\frac{x^l(PuO_2)}{x^s(PuO_2)}\right) \tag{2}$$

$$\frac{\Delta h_m(AmO_2)}{R}\left(\frac{1}{T_m(AmO_2)}-\frac{1}{T_m}\right)=\ln\left(\frac{x^l(AmO_2)}{x^s(AmO_2)}\right) \tag{3}$$

$$\frac{\Delta h_m(NpO_2)}{R}\left(\frac{1}{T_m(NpO_2)}-\frac{1}{T_m}\right)=\ln\left(\frac{x^l(NpO_2)}{x^s(NpO_2)}\right) \tag{4}$$

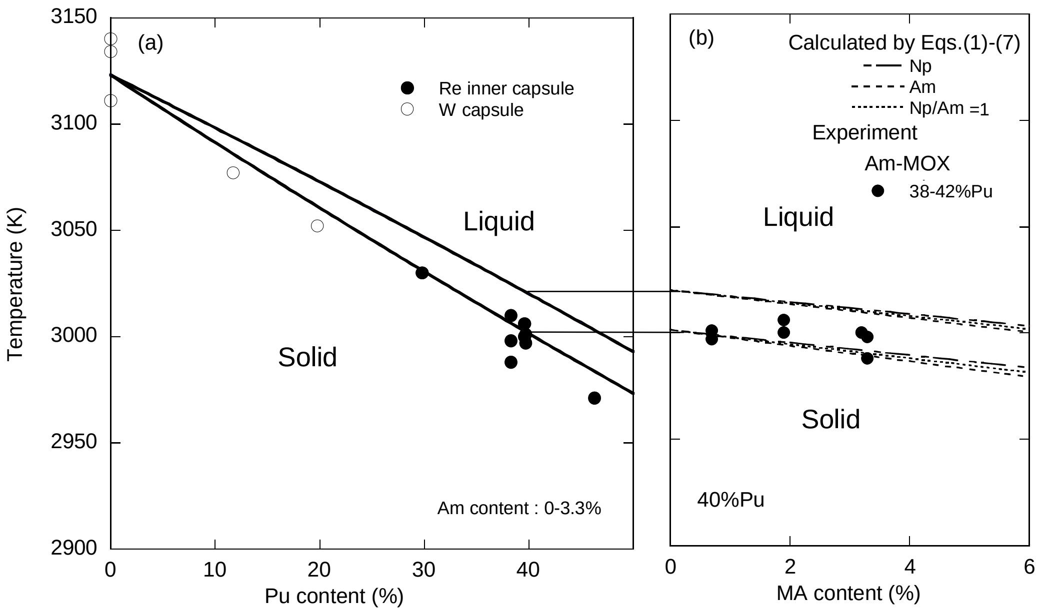
$$x^l(UO_2)+x^l(PuO_2)+x^l(AmO_2)+x^l(NpO_2)=1 \tag{5}$$

$$x^s(UO_2)+x^s(PuO_2)+x^s(AmO_2)+x^s(NpO_2)=1 \tag{6}$$

$$\Delta h_m(i)=nRT_m(i) \tag{7}$$

where T_m is the melting temperature of $(U,Pu,Am,Np)O_{2.00}$ solid solution, $T_m(i)$ and $\Delta h_m(i)$ are melting temperature and heat of fusion at $T_m(i)$, respectively, of the pure substance i, $x^l(i)$ and $x^s(i)$ represent the mole fractions of component i in the liquid and the solid solutions, respectively, and R is the gas constant. In the case of a compound of MO_2, the relation in $T_m(i)$ and $\Delta h_m(i)$ is described by Eq. (7). The melting point of UO_2, PuO_2, AmO_2 and NpO_2 were 3 123 K [8], 2 843 K [8], 2 773 K [8] and 2 833 K [23], respectively. The calculation results are shown in Figure 1. Figure 1(a) shows the variation of melting temperatures in the UO_2-PuO_2 system, and Figure 1(b) shows the melting temperature variation of $(U_{0.6-y'}Pu_{0.4}Am_{y'})O_{2.00}$, $(U_{0.6-y''}Pu_{0.4}Np_{y''})O_{2.00}$ and $(U_{0.6-2y}Pu_{0.4}Am_{y},Np_{y})O_{2.00}$. The addition of 1% Am content causes the solidus temperature to decrease about 4 K. The effect of Np is smaller than that of Am and causes about a 4 K decrease. In the case of 5% MA content, the decrease of the melting temperature is estimated to be less than 20 K. However, melting temperature of Np-containing MOX has never been measured, so it is desirable to do so.

Figure 1: The variation of solidus and liquidus temperature in (a) UO_2-PuO_2 system and (b) $(U_{0.6-z-y'-y''}Pu_{0.4}Am_{y'}Np_{y''})O_{2.00}$



Thermal conductivities

It is well-known that thermal conductivity of MOX fuel varies significantly with density, O/M ratio and temperature. Some models [24-27] for MOX have been reported, but the data are in the limited ranges of 95-90% TD and O/M ratio = 1.95-2.00 and not at high temperatures. In the case of fast reactor MOX fuel, it is necessary to get measurements in larger ranges. In addition to demonstrating the feasibility of an advanced fast reactor it is essential to investigate the effects of MA contents.

In previous works [13-17] the thermal conductivities of MOX were measured as functions of density, O/M ratio and MA content. The sample densities were controlled in the ranges of 85-95% TD.

The thermal conductivity decreased with decreasing density, and the change was represented by the Maxell-Eucken equation as follows:

$$F = (1 - p)/1 + 0.5p$$

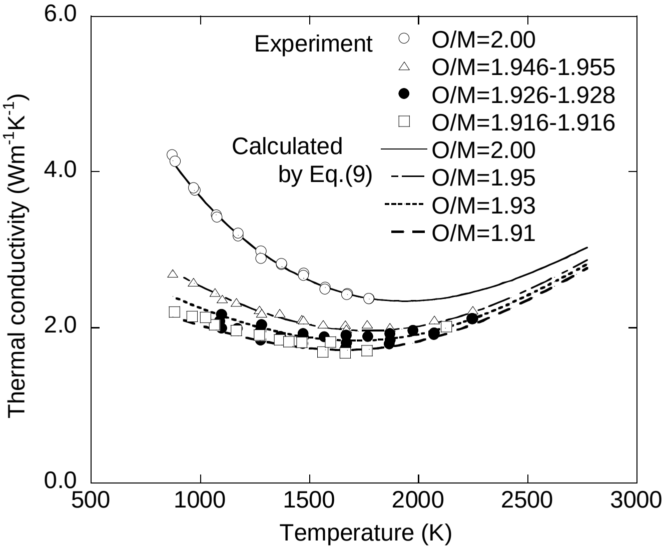
where F is the porosity correction factor and p is porosity. In the measurement as a parameter of O/M ratio, the thermal conductivity decreased significantly with decreasing O/M. The data were obtained in the O/M region of 1.90-2.00, and the classical phonon model was derived.

Figure 2 shows the thermal conductivity of 2% Am-bearing MOX. The MA contents led to a slight decrease in conductivities at temperatures less than 1 000 K. This would be a negligible change in the temperature analyses of the fast reactor fuel pellets. The following equation was derived to evaluate the thermal conductivities of fast reactor fuels from experimental data:

$$\lambda_0 = \frac{1}{\left(2.713x + 3.583\times10^{-1}\times z_1 + 6.317\times10^{-2}\times z_2 + 1.595\times10^{-2}\right) + \left(-2.625x + 2.493\right)\times10^{-4}T} + \frac{1.541\times10^{11}}{T^{5/2}}\exp\left(\frac{1.522\times10^{4}}{T}\right) \qquad (9)$$

where λ_0 is thermal conductivity of a 100% theoretical density fuel, x is deviation in $MO_{2.00-x}$, z_1 is Am content and z_2 is Np content. The calculation results are shown in Figure 2 and are in good agreement with experimental data. It is expected that Eq. (9) will be revised by expanding data in the high temperature region.

Figure 2: Thermal conductivities of $(U_{0.68}Pu_{0.3}Am_{0.02})O_{2.00-x}$ as functions of temperatures and O/M ratios



Lattice parameters

Lattice parameters are basic data for expressing material characteristics, and they are used to evaluate thermal physical properties of MOX such as thermal conductivities and heat capacities. In addition, the theoretical density of MOX has been obtained from lattice parameters and it is used to control the quality of the fuel pellets in the fabrication process. Therefore, lattice parameters are essential to develop nuclear fuels.

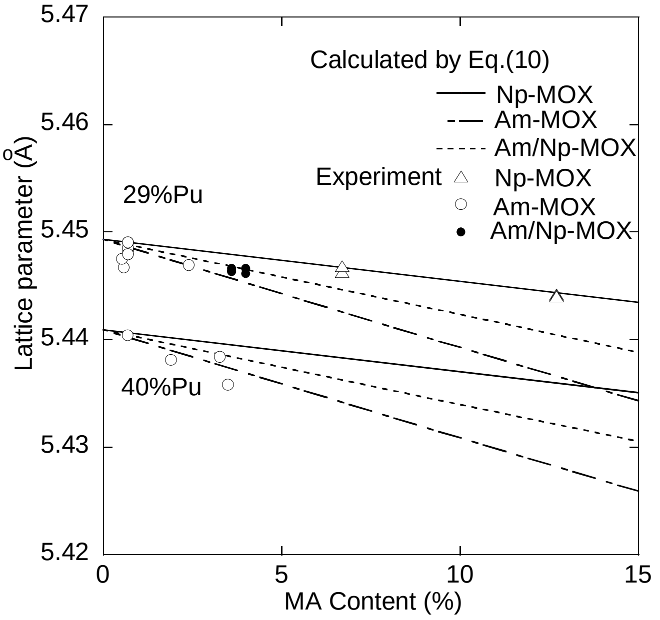
It is well-known that MOX, having a fluorite structure, is an oxygen non-stoichiometric compound which is stable in hyper- and hypo-stoichiometric compositions. The lattice parameters change significantly with Pu and MA contents and O/M ratio.

In previous works [18,19], the database for obtaining lattice parameters of (U,Pu,Am,Np)O_{2-x} was updated; there are now about 100 data points each relating to Pu, Am and Np contents and O/M ratios. The lattice parameters were analysed by using the database. The analysis results suggested that the MOX containing Np and Am was a substitutional solid solution compound. The lattice parameter of (U,Pu,Am,Np)$O_{2.00}$ obeyed Vegard's law and increased with decreasing O/M ratio. The model to calculate the lattice parameter was derived using the ionic radius model. The lattice parameters in $(U_{1-z-y'-y''}Pu_zAm_{y'}Np_{y''})O_{2.00-x}$ can be described as follows:

$$a = 4/\sqrt{3}\left[\left(r_U(1-z-y'-y'') + r_{Pu}z + r_{Am}y' + r_{Np}y''\right)(1+0.112x) + r_a\right)\right] \tag{10}$$

where r_U, r_{Pu}, r_{Am}, r_{Np} and r_a are 0.9972, 0.9642, 0.9539, 0.9805 and 1.372 Å, respectively. The model reproduced the experimental data with a standard deviation of $\sigma = \pm 0.025\%$. Figure 3 shows the calculation results in $(U_{0.7-y'-y''}Pu_{0.3}Am_{y'}Np_{y''})O_{2.00-x}$. The lattice parameters decreased with MA contents. The Am content led to a larger decrease as compared with Np content, and the figure suggested that the theoretical density of $(U_{0.7-y'}Pu_{0.3}Am_{y'})O_{2.00}$ increased from 11.10 g/cc to 11.14 g/cc with 5% Am content. The change from adding MA was very small.

Figure 3: The effects of Np and Am additions on lattice parameters of (U,Pu,Am,Np)$O_{2.00}$



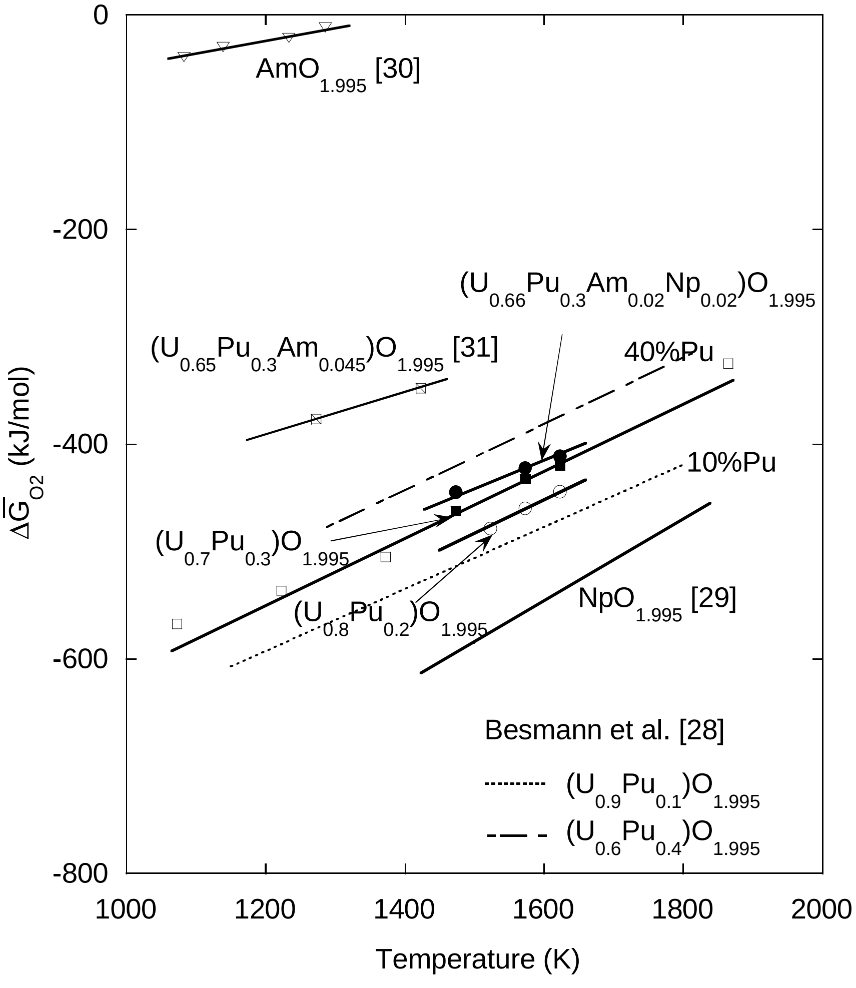
Oxygen potentials

Oxygen potentials ($\Delta\overline{G}_{O_2}$) of the MOX fuel are important data for evaluating the chemical behaviour of fuel pins. Fuel-cladding chemical interaction (FCCI) occurs on the inner surface of cladding at high burn-up and it limits the fuel lifetime. Therefore, the O/M ratio is an important parameter for controlling FCCI and determining fuel lifetime.

There have been many studies on $\Delta\overline{G}_{O_2}$ of the MOX fuels in relation to Pu content, O/M ratio and temperature [28-31]. It was reported that the $\Delta\overline{G}_{O_2}$ increases with Pu content for the MOX fuel of (U,Pu)O_{2-x} [28]. However, experimental data were scattered over ranges of more than ±100 kJ/mol, and it was difficult to understand the dependency of Pu content on $\Delta\overline{G}_{O_2}$, especially in the near stoichiometric composition range of O/M = 1.98-2.00. In previous works [20-22], the $\Delta\overline{G}_{O_2}$ of $(U_{0.7}Pu_{0.3})O_{2-x}$, $(U_{0.8}Pu_{0.2})O_{2-x}$ and $(U_{0.66}Pu_{0.3}Am_{0.02}Np_{0.02})O_{2-x}$ were accurately measured by a gas equilibrium technique.

The $\Delta\overline{G}_{O_2}$ of $(U_{0.7}Pu_{0.2})O_{2-x}$ were about 25 kJ/mol lower than those of $(U_{0.7}Pu_{0.3})O_{2-x}$. The $\Delta\overline{G}_{O_2}$ of $(U_{0.66}Pu_{0.3}Am_{0.02}Np_{0.02})O_{2-x}$ were slightly higher than those of $(U_{0.7}Pu_{0.3})O_{2-x}$. The $\Delta\overline{G}_{O_2}$ for O/M = 1.995 are plotted in Figure 4. The current data were consistent with calculated values from Besmann and Lindemer's model [28] and they well represented the effect of Pu content on the $\Delta\overline{G}_{O_2}$. The $\Delta\overline{G}_{O_2}$ of $AmO_{1.995}$ and $NpO_{1.995}$ are shown in Figure 4. The $\Delta\overline{G}_{O_2}$ of $AmO_{1.995}$ was so high compared with MOX. Therefore it seemed that the slightly higher $\Delta\overline{G}_{O_2}$ of $(U_{0.66}Pu_{0.3}Am_{0.02}Np_{0.02})O_{2-x}$ was caused by the Am addition. The difference in $\Delta\overline{G}_{O_2}$ between the two oxides was very small. It could be concluded that the effect of adding MA to the MOX fuel on FCCI was not significant.

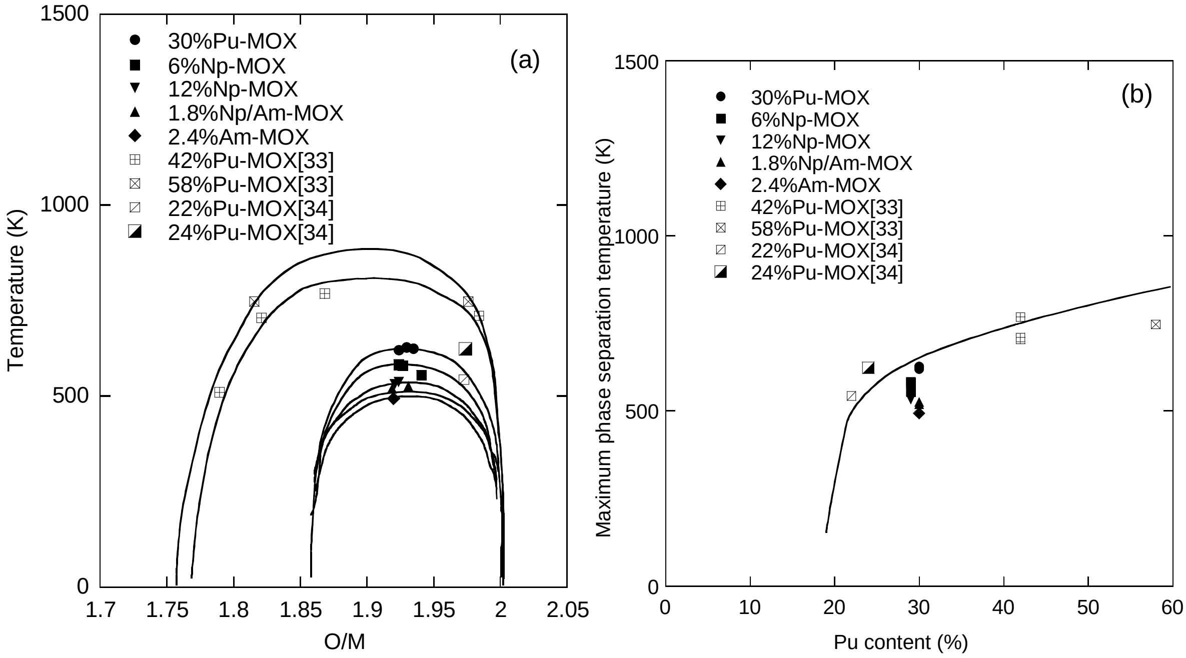
Figure 4: Comparison of the oxygen potentials of $(U,Pu,Am,Np)O_{1.995}$



Phase separation behaviour

Sari, *et al.* [32] have reported that the hypo-stoichiometric MOX has a miscibility gap above more than 20% Pu content. They gave the phase diagram of the U-Pu-O ternary system at room temperature, and noted that two fcc phases with O/M ratios of 1.85 and 1.985 exist in hypo-stoichiometric MOX with 30% Pu content. The phase separation behaviour in MA-bearing MOX has also been investigated in previous works [18,19]. The phase separation temperature of $(U_{0.7}Pu_{0.3})O_{1.92}$ was observed at 633 K, and the addition of Np and Am made it decrease as shown in Figure 5.

Figure 5: Variation of phase separation temperature depending on (a) O/M ratio and (b) Pu content



There was a concern that the separation caused microcracks in operating pellets and affected irradiation behaviour such as fission product gas release. However, the separation temperatures were lower than the coolant temperature of metallic sodium. Therefore, the phase separation would not affect the irradiation behaviour.

Conclusion

Physical properties, *i.e.* melting temperatures, thermal conductivities, lattice parameters, oxygen potentials and phase separation temperatures, of $(U,Pu,Am,Np)O_{2.00-x}$ were investigated as functions of Pu, Am and Np contents, and O/M ratios. The effects of Np and Am were evaluated, and the evaluation results are summarised as follows

1) The phase diagram of the U-Pu-O system was revised from the melting temperatures measured by a new technique using a W capsule with a Re inner liner to hold sample. The melting temperatures of fast reactor MOX fuels are 50-100 K higher than that of conventional fuels. Addition of MA decreased the melting temperature, and the decrease of the solidus temperatures was estimated to be about 20 K with 5% MA addition.
2) The additions of Am and Np caused a slight decrease in thermal conductivities at temperatures less than 1 000 K, and the effect would be negligibly small in operating temperatures of fast rector fuels. A new model was derived for calculating thermal conductivities of fast reactor oxide fuel from the measured data.
3) A model to calculate the lattice parameters of $(U,Pu,Am,Np)O_{2.00-x}$ was derived, and it was used to evaluate theoretical density. The theoretical density increased by 0.04 g/cc by adding 5% MA to MOX fuel.
4) The oxygen potential of MOX and MA-bearing MOX fuels were evaluated, and it was found that MA addition slightly increased them. The change of the oxygen potential by MA addition did not affect FCCI.
5) The effects of MA content, Pu content and O/M ratio on the phase separation temperatures were evaluated. The phase separation did not affect irradiation behaviour, because the separation temperature was lower than the coolant temperature of a fast reactor.

Physical properties of MA-bearing MOX were investigated, and the information gained has contributed to a better understanding of the effect of MA content on them. It was confirmed that MA addition did not significantly affect the fuel properties. The evaluation results of melting temperature and thermal conductivity offer a technical base for the current fuel design study of the JSFR oxide fuel.

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Minor actinide recycling in sodium fast reactor: 2008 status of Phénix experimental programme

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Abstract

The main developments in France for fast reactor fuels have been devoted to their utilisation in sodium-cooled fast reactors (SFR) and gas-cooled fast reactors (GFR). Compared to current reactors and associated with a closed fuel cycle, these future systems should allow an enhanced economic competitiveness, further enhanced safety, the use of proliferation-resistant technologies and processes, a flexible and efficient use of natural resources and a significant reduction of long-life radioactive elements in ultimate waste.

Concerning the SFR, MOX fuel remains the reference option based on the long and large experience gained in France and abroad for this kind of fuel. The main issue is the incorporation of minor actinides in the U-Pu fuel for transmutation. For long-term applications, second options are carbide, rather than metal or nitride.

Within this context, this paper reports on the status in 2008 of all transmutation tests in progress at the SFR Phénix power plant. The paper also deals with a presentation of the important end-of-life test programme which will be conducted just before the final shutdown of the reactor expected in 2009. Indeed, the Phénix reactor has industrially demonstrated that the sodium-cooled fast neutron reactor type, combined with fuel cycles implementing plutonium and minor actinides, is a crucial element to enable nuclear energy to meet the necessary criteria involved in sustainable development.

Introduction

After use, PWR fuel contains approximately 95% uranium, 1% plutonium and 4% fission products. In France, this spent fuel is reprocessed and the recovered uranium and plutonium are partially recycled in LWR, while the fission products, including minor actinides, are vitrified in ultimate waste, to be stored in a geological formation. The purpose of separation procedures is mainly to isolate the fission products that have a very long half-life and high radioactivity, so that they are no longer part of the ultimate waste whose radiotoxicity, heat and volume are so drastically reduced. Then these separated products have to be transmuted in a reactor or a dedicated tool.

In practice, most fission products are difficult to recycle. Only technetium is relatively easy to process, but as shown by the ongoing ANTICORP experiment at the Phénix reactor of CEA-Marcoule, its transmutation consumes core neutrons by capture with a low transmutation rate which is not adapted for an industrial level. Other FP (iodine, caesium), for which this procedure would be advantageous, are either difficult to separate (^{137}Cs) or difficult to process (iodine because of its high volatility). This leaves the minor actinides as the only remaining candidates. Although they constitute only 0.1% of the used fuel, their very long half-life and high activity mean that they will represent the majority of the potential radiotoxicity after a few centuries (considering that plutonium is recycled in LWR).

The transmutation of these actinides can take place efficiently only in a fast reactor where the neutrons are sufficiently available and have the energy required to cause their fission. In a LWR, captures largely exceed fissions, leading to the formation of superior actinides such as californium. These are equally inconvenient because of their long half-life and higher radioactivity than that of minor actinides. Within this context, this paper aims at illustrating the vast experimental programme that CEA has performed with the Phénix reactor, in order to demonstrate the scientific and technical feasibility of minor actinide transmutation under fast neutron flux.

Results of experimental irradiation tests in Phénix

The purpose of the French experimental programme is to assess the possibility of eliminating long-lived radioactive waste through transmutation tests in fast neutron critical reactors, such as the CEA-EDF Phénix reactor in Marcoule. Transmutation fuel development is considerably more challenging than conventional fuels and necessitates important fuel behaviour data.

Homogeneous-mode MA bearing fuels

Since the 1980s, the transmutation of the actinides neptunium, americium and curium in SFR fuel has been proposed. The Institute for Transuranium Elements (ITU, Karlsruhe), in collaboration with the CEA, launched the Superfact 1 experiment to demonstrate the feasibility of the transmutation of ^{241}Am and ^{237}Np using MOX fuel. The ITU fabricated the fuel pellets and the experimental pins while CEA performed the design basis studies, the assembly of the experimental irradiation device (capsule) and the irradiation in the Phénix reactor. The post-irradiation examinations (PIE) and the interpretations were performed jointly by the two partners. The preparation of the experiment occurred from 1984 to 1986, the irradiation from 1986 to 1988 and the PIE from 1989 to 1992.

The Superfact 1 irradiation established that the homogeneous oxide-type fuels containing either 2 wt.% ^{237}Np, or 2 wt.% ^{241}Am behave almost identically (as shown in Figure 1) to a standard fuel for a linear power between 380 W/cm and 325 W/cm and at respective burn-up of 6.6 at.% and 6.5 at.%, corresponding to transmutation rates of these minor actinides of 30%. Another important result to be noted is the total release by the fuel of the helium produced during the irradiation, as shown in Figure 2. For this transmutation ratio, the clad corrosion is the same as that of standard fuel, and the central hole is less than 1 mm shorter than for standard fuel (due to smaller linear power). The He release volume is four times greater for $(U,Pu,Am_{0.02})O_2$ pin than for standard pins. The same fuel microstructure evolution as for standard fuels is observed: U, Pu and Np radial distributions are flat, which indicates that there is no significant actinide redistribution. Finally, for low liner power, there is no real influence of the low MA content, up to a burn-up equal to 6.6 at.%, except for the He release of the Am fuel.

Figure 1: Optical micrograph of $(UPuAm_{0.02})O_2$ irradiated in a fast flux at 6.5 at.%

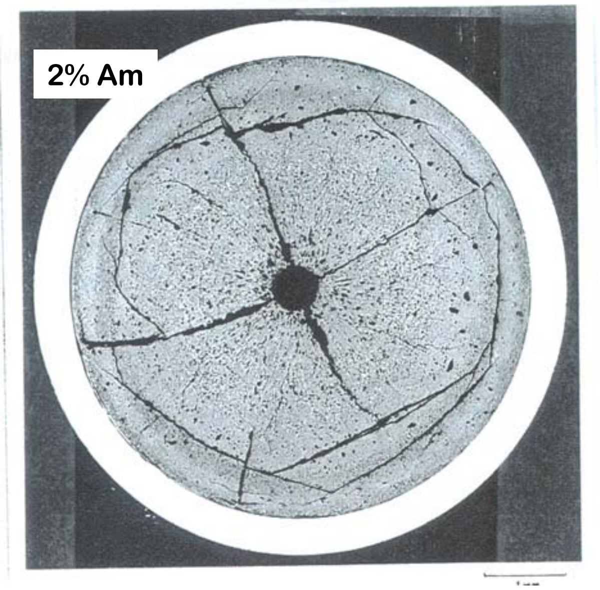
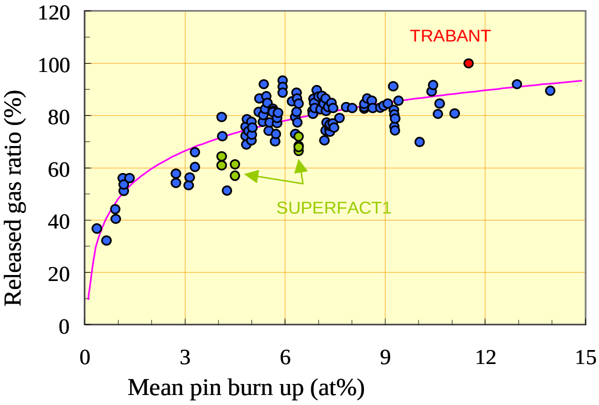


Figure 2: Released gas ratio versus fuel burn-up for Superfact and Trabant MA experiments



From these results, it is possible to assert that, thanks to the optimisation of the dimensioning of the pins (pellet-cladding space, volume of the void tank within the pin, cladding thickness), the usual performances of the fuel without minor actinides can be also obtained for a fuel containing up to a content of 2.5 wt.% americium and neptunium. Thus, the Superfact 1 experiment carried out in Phénix provides a first demonstration of the technical feasibility of the homogeneous-mode transmutation of the minor actinides in a SFR.

In a scenario where the minor actinides to be transmuted are diluted in the core fuel under the homogeneous mode, their content has to be limited to a maximal value of around 5% in SFR critical reactors due to safety considerations and reactor operation. As described above, the Superfact 1 experiment in Phénix was the early demonstration of the feasibility of this type of recycling using MOX fuel. Today, the homogeneous transmutation experiments are mainly the three Metaphix 1, 2 and 3 subassemblies containing experimental pins of metallic UPuZr fuel with dispersed minor actinides, under the framework of an agreement with the Japanese CRIEPI (Metaphix 1 was unloaded in 2004, Metaphix 2 in 2006). Of course, there are also 2 kg of americium distributed in the fuel of the standard core caused by plutonium ageing, and the satisfactory behaviour of this standard americium-bearing fuel is of interest for MA transmutation.

Minor actinide contents could reach significantly higher values in subcritical systems (ADS), and the aim of Futurix-FTA experiments (oxide, nitride and metallic ADS fuels in the same irradiation conditions) is to study the in-pile behaviour of such fuels. The experiment is planned beginning in 2007 up to the final shutdown of Phénix in 2009.

Heterogeneous-mode MA bearing fuels

An alternative way to the homogeneous recycling mode consists of a decoupled fuel/minor actinide management by loading minor actinides in an inert matrix or in an UO_2 matrix, located in radial blankets of a SFR. The major advantages of this heterogeneous recycling mode are the following:

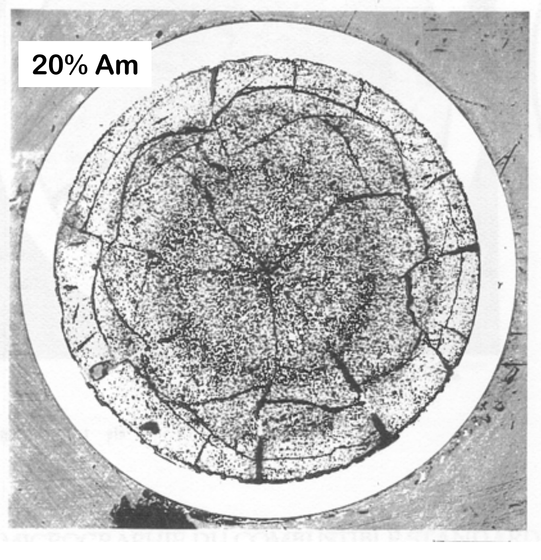
- this model has an impact of only few per cent variation of the sodium void coefficient;
- it has almost no impact on the core management;
- it enables irradiation time for minor actinides blanket longer than the one of the core fuel (typically two times), enabling a high MA transmutation ratio;
- proliferation resistance is increased compared to traditional UO_2 blankets, since introduction of actinides degrades the plutonium fissile quality.

The advantages of the concept are counterbalanced by sharp challenges that have to be carefully considered for the assembly design:

- fuel behaviour under irradiation (swelling and helium production);
- high specific heat for assembly manufacturing;
- high decay heat level for in-core and out-of-core assembly handling;
- high neutron source level at the fuel treatment step.

This first point was tackled by the Superfact 1 experiment carried out in the Phénix reactor, consisting of irradiating solid solutions of uranium and minor actinide compounds with the following contents: $(U_{0.55},Np_{0.45})O_2$ and $(U_{0.6},Am_{0.2},Np_{0.2})O_2$. Strong swelling accompanied by the formation of very high porosity was observed, as seen in Figure 3, on the irradiated fuel from a burn-up of 4.5 at.%. This important swelling is linked to the formation of numerous helium bubbles (helium being formed through the alpha decay of ^{242}Cm, coming from neutron capture of ^{241}Am), and led to an interaction between the fuel and the cladding, and finally to the mechanical deformation of the cladding.

Figure 3: Optical micrograph of $(U_{0,6},Am_{0,2},Np_{0,2})O_2$ irradiated at 4.5 at.%



Further SFR transmutation fuel development

For high MA content bearing fuels, the main concern is linked to the influence on O/M ratio: O/M of (U,Pu,Np,Am,Cm)O_{2-x} could be lower than for standard fuel. This could also correspond to a lower thermal conductivity, thus leading to a significant Am redistribution and high local concentration. Decrease of melting temperature is also a factor to be taken into account. For these reasons, the future work will have to deal with:

- fresh fuel characterisation of (U,Pu,MA)O_{2-x} samples, with several O/M ratios (from ≈1.94 to ≈1.99) and with MA amounts covering Am redistribution; measurements of O/M, partial pressures of Am, U and Pu species, thermal conductivity and melting temperatures will have to be performed;
- more detailed PIE of existing already-irradiated MA bearing fuels;
- modelling of Am redistribution laws in correlation with Pu and O/M radial redistributions, as a function of partial pressures of Am, Pu and U species;
- influence of local Am concentrations on thermo-physical properties.

With the existing results, it is thought that the development of homogeneous low MA content MOX fuel no longer necessitates a specific or heavy R&D programme, in contrast with heterogeneous high MA content UO_2 blanket fuel whose properties and irradiation behaviour have still to be largely investigated. The main aspect of the forthcoming programme for homogeneous fuel will deal with the technological feasibility of fuel fabrication, especially when curium is incorporated into the fuel, and transmutation demonstration in fast neutron reactor at pin and subassembly scale. This last item will be undertaken through the Global Actinide Recycling International Demonstration (GACID), an international collaboration programme with the United States DOE, Japan's JAEA and the French CEA, to be performed using the Monju SFR when restarted (Figure 4).

Figure 4: Irradiation programme for homogeneous MA transmutation mode

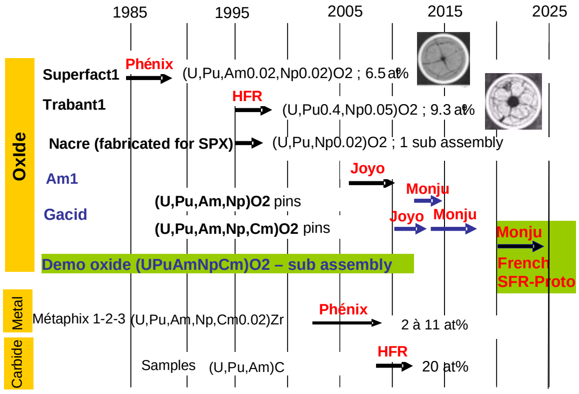
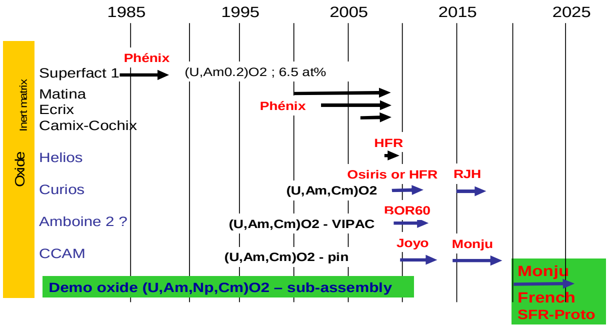


Figure 5: Irradiation programme for heterogeneous MA transmutation mode



Actual research at Phénix also deals with the possibility of heterogeneous transmutation. The objectives of the Ecrix and Camix-Cochix experiments are to test different concepts of transmutation targets (inert matrices containing particles of americium) in slightly moderated neutron spectra, either in the core (Ecrix-B), or in the fertile blankets (Ecrix-H, Camix-Cochix). Although the Ecrix-B experimental device is still in the reactor after 410 EFPD, Ecrix-H irradiation was terminated in early 2006 after having reached a fission rate of approximately 35 at.%. The pin is currently being examined in the CEA-Marcoule Irradiated Elements Cell (CEI). For the Camix and Cochix experiments, 2006 was primarily devoted to assembling the rigs at Phénix, and to processing the irradiation request with the safety authority; the irradiation test will was then started in 2007.

For the heterogeneous transmutation mode, the future programme will include analytical irradiation experiments in thermal flux reactors, such as French Osiris and RJH when started or European HFR, in parallel with transmutation demonstration at pellet then pin level in the Japanese Joyo and Monju reactors (Figure 5).

End-of-life Phénix tests

Before the final shutdown of the reactor, which is planned in 2009, an important experimental programme with specific core configurations will be devoted to a better understanding and validation of the neutron, thermal, thermo-hydraulic and safety behaviours of the reactor. Subassembly reactivity worth will be measured using substitutions of S/A in core central position and of a control rod by a gas volume. Control rod withdrawals will allow non-symmetric control rod configuration in static and dynamic modes. Precise S/A decay heat and control rod worth will be measured by different techniques. In terms of safety, demonstrations will aim at a better verification of calculation margins of SFR.

An important issue is linked to the investigation of scenarios for Phénix negative reactivity transient explanation; tests with low and zero power will produce new data in support of the possible core flowering, resulting in a very specific configuration of a moderated carrier near a blanket S/A.

Conclusion

In conclusion, the international context is favourable to the renaissance of nuclear energy, among other energy sources, and the Generation IV International Forum has thus far been a very active framework to define the objectives for nuclear systems after 2040 and to prepare the development of enabling key technologies with regard to international co-operation. Generation IV fast neutron systems are considered as key components for the renewal of the French generating facilities after 2040, and for the management of the transuranium elements generated by the current power reactors. In France, transuranium element transmutation has been studied in detail thanks to the extensive experimental irradiation programme carried out, in large part, with tests in the fast flux Phénix reactor for which power restart has been effective since late 2003. Flux conditions of Phénix are well suited to allow the studies of irradiation damages of fuels and targets for transmutation under representative conditions of fast and partly moderated flux, which are considered to be the most efficient for transmutation of minor actinides and some long-lived fission products.

A new waste management law was passed by the French parliament on 28 June 2006, demanding that P&T research continue in strong connection to Gen-IV systems development and allowing a detailed cost/benefit analysis by 2012, in order to decide or not the industrial implementation of P&T, if proved of interest with regard to the consequent simplification of the conditions of the necessary geological disposal. The introduction of fast reactors, in association with the corresponding fuel cycles (U and Pu reprocessing, and separation of the minor actinides), places the entire nuclear energy sector in a context of sustainable development, both in terms of resource management and in terms of waste management; the Phénix sodium fast reactor has contributed the first industrial demonstrations in this field, first in the 1980s for breeding, followed by similar demonstrations of transmutation since 2003. In this context, it is considered that Phénix has already brought outstanding results in support of the feasibility of MA transmutation under representative conditions of future SFR.

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Session III

Progress in partitioning, waste forms and management

Chairs: C. Hill, T. Inoue

Future nuclear fuel cycles: Prospects and challenges

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Abstract

Both in France and world wide, nuclear power has the potential to curtail the dependence on fossil fuels and thereby to reduce the amount of greenhouse gas emissions while promoting energy independence. The global energy context pleads in favour of a sustainable development of nuclear energy since the demand for energy will likely increase, whereas resources will tend to get scarcer and the prospect of global warming will drive down the consumption of fossil fuel. Therefore, retaining nuclear power as a key piece of the nation's energy portfolio strengthens French energy security and environmental quality.

How we deal with nuclear radioactive waste is crucial in this context. The public's concern regarding long-term waste management led the French government to prepare and pass the 1991 and 2006 Acts, requesting in particular the study of applicable solutions for further minimising the quantity and the hazardousness of final waste. This necessitates high active long-life element [such as the minor actinides (MA)] recycling, since the results of fuel cycle R&D could significantly change the challenges for the storage of nuclear waste. HALL recycling can reduce the heat load and the half-life of most of the waste to be buried to a couple of hundred years, overcoming the concerns of the public related to the long life of the waste thus aiding the "burying approach" in securing a "broadly agreed political consensus" of waste disposal in a geological repository. It appears clearly that long-lasting nuclear options will include actinide recycling.

Within this framework, this paper presents the progress obtained at CEA Marcoule on the development of innovative actinide partitioning hydrometallurgical processes in support of their recycling under different still-open options, either in homogeneous mode (MA are recycled at low concentration in all the standard reactor fuel) or in heterogeneous mode (MA are recycled at higher concentration in specific targets, at the periphery of the reactor core). Recovery performances obtained on a recent test in high active conditions using the Ganex process (grouped actinide separation connected to homogeneous recycling) are presented and discussed, as compared to the demands of P&T scenarios. New results c also oncern major improvements and possible simplifications of the Diamex-Sanex process, whose technical feasibility was already demonstrated in 2005 for americium and curium partitioning (heterogeneous mode).

In the coming years, next steps will involve both better in-depth understanding of the scientific basis of these actinide recycling processes, and for the new promising concepts, the studies necessary prior to industrial implementation of these processes towards the 2012 milestone.

1 Introduction

After use, PWR fuel contains approximately 95% uranium, 1% plutonium and 4% fission products. In France, this spent fuel is reprocessed and the recovered uranium and plutonium are partially recycled in LWR, while the fission products including minor actinides are vitrified in ultimate waste, to be stored in a geological formation. The purpose of separation procedures is mainly to isolate the fission products that have a very long half-life and high radioactivity, so that they are no longer part of the ultimate waste whose radiotoxicity, heat and volume are so drastically reduced. Then these separated products have to be transmuted in a reactor or a dedicated tool.

As mentioned, the recycling of the plutonium is essential to reduce the potential long-term radiotoxicity of the ultimate wastes, but we can look to further reduce it by an additional recycling of minor actinides. During recent years, major progress has been achieved on MA partitioning thanks to the studies performed at the CEA Atalante at Marcoule [1,2]. Specific extractant molecules have been successfully developed and tested, and in 2005 the demonstration of the technical feasibility of the Diamex-Sanex process for americium and curium selective extraction was demonstrated using the hot cells of the facility [3,4]. This liquid-liquid extraction experiment carried out on nearly 15 kg of spent nuclear fuel showed the efficiency of the recovery process for recycling neptunium, americium and curium with rates higher than 99%. Since then, regarding future fuel cycles, most of the research in France has been conducted in the framework of the 28 June 2006 Waste Management Act [5] which defines a roadmap and the main objectives to be achieved in a near future, towards the industrial potentialities of the diverse P&T options by 2012 and the construction of a prototype reactor for MA transmutation demonstration by 2020. Clearly, these objectives consist of:

- defining the several options of interest, which could be successively deployed (all actinide, americium only, heterogeneous, homogeneous,…);
- assessing benefits/costs ratio for the several recycling options, considering different criteria and “densification” of the final storage;
- designing and optimising the separation processes, transmutation fuels and their fabrication process;
- gathering technical elements for industrial operation evaluation.

The aim of this paper is to provide an overview of the experimental results obtained since 2006 within the research programmes dealing with optimising the concept and further increasing the compactness of operations of an enhanced partitioning process, such as the Diamex-Sanex for heterogeneous recycling. The paper also presents first test results carried out in 2008 on the Ganex process adapted to the possible recycling strategy of all transuranic elements together. Finally, for new promising concepts and corresponding to the milestone objectives of the 2006 Act, the consolidation of future studies which are needed prior to potential industrial implementation of these processes are described.

2 Recent progress for partitioning process

2.1 Improvement of the Sanex process

Recycling trivalent minor actinides by separation has been investigated at CEA Marcoule over the last two decades using the Sanex process with ligands containing soft donor atoms such as the tridentate nitrogen bearing TriPyridylTriaZine (TPTZ) and BisTriazinylPyridines (BTP). But these systems could only be implemented as second cycles after the Diamex process that coextracts first trivalent 4f and 5f elements [respectively Ln (III) and An (III)] and, in order to develop simpler one-cycle An(III) partitioning processes, new extracting agents are still under investigation at CEA Marcoule. This is the case of the TetraOctyl-DiGlycolAmide (Todga) which presents a very high extraction affinity towards trivalent actinides.

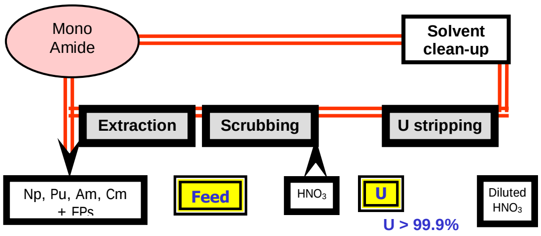
In this context, CEA researchers have imagined a one-cycle An(III)/Ln(III) separation process based on the Todga/TBP solvent, in view of the fact that it provides such a high extraction affinity toward trivalent 4f and 5f elements, that the selective stripping of the An(III) appears feasible by a hydrophilic polyaminocarboxylate complexing agent in a buffered solution, while the Ln(III) remain complexed by Todga in the organic solvent thanks to a salting-out agent. The major difficulty encountered was to minimise as much as possible the impact of the solvent acidity (nitric acid is mostly extracted by TBP) on the pH of the aqueous stripping solution and hence on the An(III)/Ln(III) separation yields. Various hydrophilic complexing agents have been investigated in combination with various carboxylic buffers. The choice ended up with diethylenetriamine-pentaacetic acid (DTPA) and malonic acid, the concentrations of which have been optimised respectively to complex all the An(III) potentially present in the Purex raffinate and to buffer the aqueous stripping solution.

Three tests have been performed by varying the pH in the An stripping section, and good results were obtained during these counter-current inactive and spiked tests performed in 2008 at CEA Marcoule with a satisfactory comparison between experimental and calculated concentration profiles. Results of a hot Sanex-Todga test, to be performed at the Atalante facility in 2009, should confirm the process performances with recovery yields of An(III) larger than 99.9% with high decontamination factors towards lanthanides(III).

2.2 *Development of the Ganex process*

The Ganex process aims at the recovery of all transuranium elements from the HA spent fuel dissolution raffinate. It consists of two steps: partitioning of uranium alone (Figure 1), then partitioning of actinides from fission products and lanthanides.

Figure 1: Partitioning of uranium from the HA spent fuel solution



Among the different processes developed for the partitioning of trivalent minor actinides, the Diamex-Sanex process using the HDEHP-DMDOHEMA solvent was selected in 2006 as the best candidate to achieve the group actinide separation. This process is able to separate americium and curium from the lanthanides and the other fission products, but it had to be modified in order to take into account the presence of neptunium and plutonium along with the trivalent minor actinides within the Ganex process. For the first step, a counter-current test performed in 2008 reached the objective of the Ganex first cycle of recovering more than 99.99% of the total amount of uranium. This test confirmed the good performance of DEHiBA and N,N-dialkylamide for the extraction of uranium from nitric acid and its selectivity towards the fission products. The model based on the acquisition of experimental distribution data with corrections for the activity coefficients of the components in the aqueous phase was verified by the good agreement observed between the experimental and calculated profiles.

After demonstrating this good possibility to quantitatively separate Np, Pu, Am and Cm altogether, batch experimental data were acquired to develop physicochemical models first validated on previous tests performed on An(III)/Ln(III) separation, and before being used to design the flow sheets for the cold and the hot Ganex second cycle tests. Several cold tests have been carried out on lanthanide and major fission products in order to check the hydrodynamic behaviour and the extraction performances of lanthanides, Mo, Pd, Zr, and Fe. Then, the actinide group separation was performed in four sections of laboratory scale mixer-settlers corresponding to the extraction-scrubbing, the Mo-Tc stripping, the actinide stripping (including eight stages for lanthanide scrubbing) and the lanthanide-zirconium-iron stripping steps. The contactor set up consisted of five banks of mixer-settlers of 12, 8, 8+12 and 8 stages, respectively. In terms of results, neptunium, plutonium, americium and curium were recovered all together in one liquid flow (actinide product) and the losses of transuranium in the different outputs and in the solvent were estimated at a value lower than 0.5% (neptunium essentially) at the end of the test, corresponding to a recovery yield of actinides higher than 99.5%. Nevertheless, the decontamination factors versus some lanthanides (especially Nd, Sm and Eu) were much lower than expected and the mass of lanthanides in the actinide product was around 5% at the end of the test, which was explained by an accumulation of lanthanides in the actinide stripping section which could not be resorbed during the duration of the test. The decontamination factors versus some lanthanides were not as high as expected but could be well simulated by the model.

3 Consolidation programme prior to industrialisation

Substantial developments are still necessary when considering industrial use of partitioning in the future. First of all, the research should continue along the same lines as that carried out over the past years which have proven the technical feasibility of the processes but, in parallel, the industrial aim must now be defined and highlighted, and then stated in detail through a specifications sheet laying down the performance expected, so as to target the action needed to extend the developments.

In this context, the CEA has defined a three-year programme (2010-2012) aimed at the consolidation of the innovative MA recycling processes under technical demonstration in order to gain their future industrial feasibility. The demonstration of the technical feasibility of minor actinide partitioning should be followed by tests meant to optimise the processes according to the industrial aim.

Based on the results of experiments associated with modelling, this programme will allow for the assessment of the process performance in various operating conditions. In this context, the nature of certain reagents or the formulation of extraction molecules could, if appropriate, be modified in order for example to reduce supply costs or bypass difficulties in effluent management. The endurance of solvent recycling operations is a key aspect to be guaranteed in view of industrialising the process. Testing solvent endurance and recycling operations have already been undertaken at CEA, and will continue in order to propose, if necessary, specific regeneration processes.

The dimensioning of most conventional extraction equipment used in the enhanced partitioning processes appears possible, while qualification tests on large mock-ups could be avoided when using the best known reprocessing technologies, such as those of the La Hague plant. Nevertheless, some operations, particularly those associated with solvent recycling, could require the development of specific contactors.

The consolidation study will also contain interdependent work areas. Science and engineering support is one of the areas in order to generate data to help steer process design parameters. Other domains will deal with process design parameters and flow sheet in terms of feeds, products, decontamination factors, flow rates, process losses as a function of the selected technology through possibly modelling. This work should aim to develop design objectives for the solvent, quantified in terms of process parameters such as required solvent lifetime (radiolysis and hydrolysis) and inventory, ideal distribution values and equilibration kinetics, scrubbing and stripping characteristics.

The programme is planned to deal with the characterisations of the recovered materials produced by the partitioning process; it is essential to assess the feasibility of the co-conversion of the MA solution for producing the MA oxide compound which will be the precursor of the transmutation MA-bearing fuel. For instance, the influence of the level of possible remaining impurities, such as some lanthanides, in the partitioned solution will be checked upon the fuel fabrication process parameters.

Finally, part of the programme will contribute to system studies with the objectives to consider and agree the achievements of possible global fuel cycle scenarios, in a correct coherency with the process performances.

4 Conclusion

P&T technologies allow meeting the objectives of countries in Europe and abroad which have decided upon or envisage different nuclear power and fuel cycle policies. For countries favouring the continuity of the nuclear energy development towards the deployment of new fast spectrum systems, one of the main priorities for such future fast spectrum systems is the minimisation of waste which would necessitate the recycling of actinides and therefore implementation of their partitioning (specifically or grouped).

Important results have already been obtained at the CEA Atalante facility both for homogeneous and heterogeneous recycling, dealing with some simplified flow sheets such as the Ganex or the Sanex-Todga processes. In case the strategy of americium alone recycling would be selected, the process definition for "direct" recovery of this actinide must also be studied in order to propose an effective flow sheet to be carried out as a future hot test in the Atalante facility.

All these aqueous processes have been brought to a point where there is reasonable assurance that industrial deployment can be successful. Nevertheless, future R&D work will have to cope with the objective of such a potential industrialisation, which is the purpose of the consolidation programme launched by CEA to be implemented from 2010 to 2012 within the new Waste Management Law passed by the French parliament on 28 June 2006, demanding that P&T research will continue in close connection to Gen-IV systems development and allowing a detailed cost/benefit analysis by 2012, in order to determine whether or not to implement P&T at the industrial scale, if proved of interest with regard to the consequent simplification of the conditions of the necessary geological disposal.

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ACSEPT, a new step in the future demonstration of advanced fuel processing

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Abstract

Actinide recycling by separation and transmutation is considered world wide and particularly in several European countries as one of the most promising strategies to reduce the inventory of radioactive waste, thus contributing to making nuclear energy sustainable. By joining together European universities, nuclear research bodies and major industrial players in a multi-disciplinary consortium, the FP7 EURATOM Fission Project ACSEPT provides the sound basis and fundamental improvements for future demonstrations of fuel treatment in strong connection with fuel fabrication techniques. In accordance with the Strategic Research Agenda of the Sustainable Nuclear Energy Technology Platform (SNE-TP), the timelines of this four-year R&D project (2008-2012) should allow the offering of technical solutions in terms of separation process that may be reviewed by governments, European utilities as well as technology providers at that time horizon. By showing a technically feasible recycling of actinides strategy, ACSEPT will certainly produce positive arguments in the sense that European decision makers, and more globally public opinion, could be convinced that technical solutions for a better management of nuclear wastes are now technologically feasible.

ACSEPT is thus an essential contribution to the demonstration, in the long term, of the potential benefits of actinide recycling to minimise the burden on geological repositories. To succeed, ACSEPT is organised in three technical domains: i) Considering technically mature aqueous separation processes, ACSEPT will optimise and select the most promising ones dedicated either to actinide partitioning or to group actinide separation. These developments are appropriately balanced with an exploratory research focused on the design of new molecules. ii) Concerning pyrochemical separation processes, ACSEPT first focuses on the enhancement of the two reference cores of process selected within EUROPART. R&D efforts shall also be brought to key scientific and technical points compulsory for building a whole separation process. iii) All experimental results will be integrated by carrying out engineering and systems studies on hydro- and pyroprocesses to prepare for future demonstration at a pilot level. In parallel, with a view to consolidate future actinide recycling strategies, ACSEPT is in charge of the design of the minor-actinide-containing pins, prior to their fabrication in the future FAIRFUELS FP7 project.

A training and education programme is also being implemented to share the knowledge within the partitioning community and present and future generations of researchers. Specific attention will be given to the funding of multi-partite post-doctorate fellowships.

Introduction

When considering sustainable energy development world wide, one of the challenges with regard to nuclear energy is the minimisation of the production of long-lived radioactive waste but also the optimisation of the use of natural resources with an increased resistance to proliferation. Partitioning and transmutation (P&T), associated to a multi-recycling of all transuranics (TRU), should play a key role in the development of a sustainable nuclear energy through Gen-IV fast reactor systems associated with a closed cycle.

Therefore, in parallel with the development of fast neutron reactors, a separation and treatment strategy needs to be implemented, aiming at a closed fuel cycle that includes the recycling of actinides. This strategy will in particular have to comply with the world-wide policy of safeguarding and proliferation resistance that favours recycling modes with co-management of actinides. It would also permit the transition from the currently practiced mono-recycling of plutonium in light water reactors (LWR) to actinide (U, Pu, MA) recycling in Gen-IV reactors, thereby allowing minimisation of radiotoxicity in the ultimate waste.

Considering this evolution towards closed fuel cycles, it has been considered as a priority to strengthen the links and synergies with transmutation as well as with geological disposal or interim storage activities. This evolution towards more integration should thus materialise and be consistent with the building of a European vision on P&T and more globally on future sustainable nuclear systems, such as those considered within the European Sustainable Nuclear Energy Technology Platform (SNE-TP).

Despite the different national strategies envisaged for managing nuclear wastes, the options for actinide recycling implementation show a significant common trunk which allows drawing a consensual European roadmap for research and development activities as well as for future pilot-scale fuel cycle facilities. Actually, P&T has been initially pointed out in numerous studies as the strategy that can relax the constraints on geological disposal, and reduce the monitoring period to technological and manageable time scales. Moreover, even when considering the phase-out of nuclear energy, the combination of P&T and dedicated burner such as ADS technologies, but this time at regional scale, would allow meeting the objectives of minimising the long-lived waste to be ultimately disposed of.

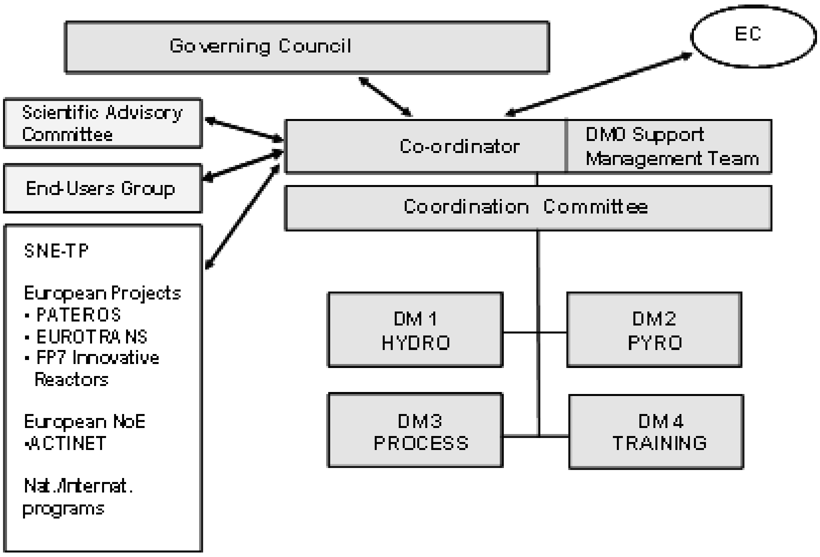
To implement this strategy at the horizon 2040-2050, it is expected around 2012 to review national positions, as well as the impact of the recycling of actinides on geological repository in terms of requirements and capacity. Scenarios for the recycling of actinides implementation and the evaluation of options such as homogeneous/heterogeneous recycling should also take into account the national capacities in fuel reprocessing and fuel fabrication, notably during the transient period. A review of group recovery of transuranium elements (TRU) vs. minor actinides (MA) selective separation should also be undertaken by 2012. Priorities should also be given following the review of ADS vs. critical fast systems potentialities and their different coolants. All these reviews should lead to take decisions on demonstration facilities to be built at a time horizon 2015-2020.

In line with these time scales, the FP7 ACSEPT Collaborative Project (2008-2012) provides a structured R&D framework with the ambitious objective to develop chemical separation processes compatible with fuel fabrication techniques, in view of their future demonstration at the pilot level, so as to offer technical solutions to the various fuel cycle options envisaged today. Flexibility will thus be kept as a main driving force while waiting for the consolidation of these options.

To optimise the production, dissemination and potential exploitation of the generated knowledge, ACSEPT is structured in three technical domains associated to clearly identified issues, plus a forth domain fully dedicating to training and education.

The ACSEPT multidisciplinary consortium is composed of 34 partners (from 12 Europeans countries plus Australia and Japan) from European universities, nuclear research bodies and major industrial players.

Figure 1: ACSEPT organisational chart



Technical programme

The separation of all significant transuranium (TRU) elements for recycling in fast neutron reactors or burning in advanced-driven systems is a key feature of advanced fuel cycles. However, new reprocessing technologies (spent fuel dissolution, actinide separation and fuel refabrication) are required to address the challenges, which cannot be met by the current generation of chemical plants. Two strategies are proposed for the recycling of the actinides issuing from the various forms of future nuclear fuels (oxides, carbides and nitrides or metallic fuels): *i)* their homogeneous recycling in mixed fuels (via a prior group separation of the actinides: GANEX concept); *ii)* their heterogeneous recycling in targets or core blankets (via their selective separation from fission products).

Two major technologies have been explored so far to meet these challenges:

- hydrometallurgical processes that benefit from more than 60 years of research and developments and a long-lasting proven experience at the industrial level;
- pyrochemical processes first studied in the 50-60s for the treatment of spent fuel from molten salt reactors and breeder reactors and more recently, with a renewed interest at the end of the 80s, for specific applications, but without reaching the industrial development level.

Hydrometallurgy

As regards partitioning processes, many countries have developed hydrometallurgical processes over the past four decades to recover TRU elements in order to decrease the radiotoxic inventories of nuclear waste. However, none of these processes has ever been implemented at the industrial scale, though their R&D have sometimes reached demonstration tests at the laboratory scale. Most of the partitioning strategies rely on a three-step approach:

- separation of U (and sometimes also Pu) from spent fuel dissolution liquors;
- An(III) + Ln(III) co-extraction;
- An(III)/Ln(III) separation.

The latter step is considered the most difficult because of the similar chemical properties of 4f and 5f elements.

The processes developed around the world differ from the extracting systems involved in these different steps and the possibility to merge two of these steps in a single one.

For the first step, TBP is the basis of the PUREX, UREX and COEX™ processes, developed in Europe and the US, whereas monoamides support the BAMA process developed in Japan and India. As these processes are well advanced in these countries, ACSEPT does not intend to invest any resources in this topic.

For the second step, malonamides, CMPO and TODGA are used in the DIAMEX, TRUEX and ARTIST processes, respectively developed in Europe, US and Japan. TRPO (trialkyl-phosphine oxide) developed in China or UNEX (mixture of several extractants) jointly developed in Russia and the US allow all actinides from U to Cm to be co-extracted from the dissolution liquors. The baseline of ACSEPT for this topic is clearly the milestones reached in the FP6 IP EUROPART [1] project with the demonstration of the scientific viability of the An(III)+Ln(III) co-extraction using the TODGA extractant on a PUREX raffinate. All the progress in terms of optimisation of the process will be measured against this baseline.

Finally, for the third step, a number of soft donor atoms containing extractants, such as polyaromatic nitrogen ligands [BT(B)P] or dithiophosphinic acids have been developed in Europe, China and Japan to selectively extract the trivalent actinides, whereas other soft donor atoms containing ligands, such as polyaminocarboxylates (HEDTA, DTPA) have been tested in France, the US (TALSPEAK) and Japan (SETFICS) to selectively strip the trivalent actinides.

The current BTBP reference system developed during FP6 IP EUROPART [2] for An(III) selective extraction does not fulfil all process development requirements but may be considered as the baseline where the project work could start from. Progress must be made in the following area: ligand stability vs. radiolysis, An(III) extraction/stripping kinetics and process simplification (3/2/1 steps).

In parallel, a new approach is investigated: the selective stripping of trivalent minor actinides after their extraction by approved lipophilic ligands (*e.g.* malonamides, DGA). The baseline clearly comes from achievements of France, USA and Japan as this concept has never been studied before in the previous FP. It thus represents an alternative route if the An(III) selective extraction cannot be developed.

The TRU actinide group separation (*i.e.* Np, Pu, Am-Cf) from spent fuel dissolution liquors (probably after a bulk U partitioning cycle) has only been scarcely investigated in previous projects, and many challenging technological obstacles (*e.g.* solvent loading capacity, oxalate substitute) have to be overcome before demonstration tests can successfully be carried out.

Whereas chemical separation processes represent a common trunk of operations that are relatively independent of the nature and type of fuel treated, head-end operations, conversion of actinides from liquid to solid state are at the interface with the fuel, either for its fabrication or its treatment once used. Dissolution and conversion are fuel-dependent operations. In Domain 1 (DM1) dissolution studies will mainly focus on fuels dedicated to the actinide homogeneous recycling.

Although current generations of fuel forms are generally oxides, future fuels may be manufactured as oxide, carbide, nitride or metallic form. Features that all the future fuels envisaged for homogeneous recycling have in common is that the Pu and minor actinide content (respectively ~20 wt.% and 5 wt.%) will be higher than that normally encountered in current oxide fuels. The burn-up, and hence heat load and activity, of these fuels will also be much higher and will present new challenges in fuel handling and processing. In addition, the matrix and Pu content of the fuels may result in increased levels of Pu-rich insolubles, however, losses of TRU to residues must be minimised to maximise the benefits gained from P&T fuel cycles.

The co-conversion of variously mixed actinide aqueous products into oxide, carbide or nitride powders, as starting materials for new fuels or targets will be the end-step to close the loop. The objectives of the research to be carried out in this field are related to the development of methods for the co-conversion of actinide solutions (variously mixed) to polyactinide containing solids for fuel preparation (Gen-IV, transmutation). To reach these objectives, different co-conversion processes have been identified, such as co-precipitation and sol-gel routes. In addition, new alternative routes such as co-denitration and co-conversion by impregnating solid extractants, followed by thermal treatment, also show promising assets but have been less studied so far. For all these processes, a common feature is the complexity of the initial mixed actinide containing solution and there is a need to cover basic studies to understand and master the co-conversion processes.

The main objectives of DM 1 is thus to develop aqueous chemical processes in the various fields of spent fuel dissolution treatment, separation process development and fuel refabrication, and to demonstrate by the end of the project their technical feasibility at the laboratory scale on actinide solutions.

Pyrometallurgy

Pyrometallurgy has been studied as an alternative strategy in the reprocessing of spent fuel for more than 50 years. Indeed, it is considered as the reference route for molten salt reactor fuel reprocessing, the reprocessing of some types of fuels today envisaged for Gen-IV might not be compatible with current hydrometallurgical processes. In FP5 PYROREP and in FP6 EUROPART projects, European scientists involved in the treatment of nuclear wastes, have carried out studies and research programmes together to increase the level of knowledge in pyrometallurgy with respect to process development, specific waste treatment and confinement.

A pyrochemical process currently includes several basic steps: separation of the fuel from the cladding material; dissolution of the material in a molten salt (except in the case of the electrorefining where no dissolution step is necessary); chemical or electrochemical reduction of oxide to metal, separation of the desired elements by electrowinning, transfer to an immiscible liquid metal phase or selective precipitation. These processes include not only a core processes but also numerous ancillary operations: head-end steps, conversion of final products to ensure compatibility with subsequent refabrication techniques, decontamination of salt and metal fluxes prior to recycling and conditioning of specific waste materials in accordance with their chemical and radiological properties.

In PYROREP (FP5) and EUROPART, the effort was put on basic data acquisition mainly in molten chloride and on core processes assessment. Two promising core processes were developed and assessed: electrorefining on solid aluminium in molten chloride and liquid-liquid reductive extraction in molten fluoride/liquid aluminium. In parallel, progress was made in the decontamination of spent chloride salt by fission products precipitation or filtration. Some original confinement matrices were also studied for waste conditioning. Finally, integration studies have been initiated in order to assess and to compare some selected process flow sheets and possibly to redirect R&D programmes. These elements clearly represent the baseline against which progress and improvements will be measured.

Consequently, important technological blocks of tasks were identified as key scientific points to be studied in ACSEPT: head-end steps must be developed in fluoride and optimised in chloride. Some ancillary steps of the core process must be assessed (exhaustive electrolysis in chloride, actinide back extraction from aluminium). The need for an electrochemical process in molten fluoride as an alternative route has also been pointed out and important efforts will be devoted to this route. Progress is still expected in salt recycling (optimisation in chloride and development in fluoride) and specific waste conditioning (establishment of reference routes).

All these activities concentrated in Domain 2 (DM2) will allow making significant advances beyond the current state of the art in pyrochemical separation processes They will also bring to DM3 lab-scale tested technological blocks of tasks that will permit the elaboration of validated reprocessing schemes that could finally lead, in future FP, to integral experiments and ultimate lab-scale demonstrations on genuine spent fuel. This work takes into account the diversity of the possible fuels of ADS or Gen-IV reactors, with a view to future demonstration at a pilot level at a longer term than the current hydrometallurgical processes. All the above expected scientific and technical advances will be measured by passing successfully different milestones (see the list).

Process and integration

Over successive European Framework Programmes novel partitioning technologies have broadly been demonstrated to be technically feasible at the laboratory scale with some current demonstrations performed with irradiated spent fuel. There are, however, significant challenges to tackle in turning the basic research and development into a deployable process at the pilot plant scale. Furthermore, there are challenges to face in process development and in integration of technology within the nuclear fuel cycle. The engineering studies proposed in ACSEPT will deploy tools which will enable early feedback and guidance to lab-scale studies. These feedback mechanisms can be used to positively select candidate separation processes for focused development, to refine the axis of research, and more generally to provide focus and rationalisation to the field of research. It thus directly benefits the development and future implementation of the technologies, incorporating and using input from all contributing partners. This approach has been for the first time implemented and successively tested on pyrochemical processes within EUROPART and will be extensively applied within DM3. Technical seminars for and by experts will be organised in the first year of the project to spread the knowledge among researchers and guarantee a good understanding and appropriation of the separation criteria to be fulfilled prior to developing processes.

Two tasks of ACSEPT will be devoted to emphasising integration between partitioning and transmutation:

- Design the fabrication technology beyond FP6 by the technologically very demanding fabrication and irradiation of minor actinide-containing pellets to be done in the FP7 FAIRFUELS Project.
- Inert matrix fuels and targets, specifically designed for the purpose of minor actinide transmutation, are significantly different in composition compared to ordinary reactor-oxide or mixed-oxide fuels. Several different types of targets and fuels are under investigation in FP6 programmes. The reprocessing aspects of the irradiated actinide targets have not been systematically studied before and ACSEPT thus proposes to address these challenges. ACSEPT contribution in this field will be limited to “paper” studies to evaluate, rank different types of targets with regard to their reprocessing capability and outline the potential key scientific issues that may be further studied in a future experimental programme.

Training and mobility, dissemination of knowledge

The ACSEPT project will seek to advance the integration of European education and training in the field of separation techniques, and actinide chemistry in particular, to combat the decline in student numbers, teaching establishments, young researchers thus providing the necessary competence and expertise for a sustainable development of nuclear energy.

DM4 will seek to improve not only motivations and skills of people but also make knowledge in the field of nuclear chemistry more transparent, improving the teaching methodology and providing an infrastructure which will support co-operative work among the members of the ACSEPT nuclear community. Of course strong links will be established with the ongoing ACTINET NoE direct or derived actions or with any post-ACTINET initiative. Complementarities rather than duplication will be obviously sought so as to optimise EC funds.

The overall goal of education and training programme in ACSEPT is to enhance the knowledge in separation sciences within the participating community. A variety of skills and equipment exist within the consortium and thus cross-cutting education is one of the better forms of teaching. In addition, researchers outside the direct community should be given the possibility to learn from the experiences gained within ACSEPT. Therefore the main activities and budget allocation will rely on the mobility of personnel.

Conclusions

With the technical advances expected within ACSEPT, advanced closed fuel cycles that include the recycling of actinides could be proposed to governments, European utilities and technology providers. A technically feasible recycling of actinides strategy will certainly produce positive arguments in the sense that:

- European decision makers and more globally public opinion could be convinced that sound technical solutions for a better management of the nuclear wastes are now technologically feasible.
- One of the identified major difficulties for nuclear energy production, *i.e.* to guarantee the non-dissemination of hazardous radionuclides within the biosphere in the far future, is on the way to be solved, thus paving the way towards sustainability.

Acknowledgements

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In memoriam

This paper is dedicated to Professor Charles Madic, who passed away on 1 March 2008.

Charles Madic began working at the CEA in 1969 in the radiochemistry laboratories located at Fontenay-aux-Roses. He then led the Chemistry Group before being appointed deputy head of the Process and Modelling Unit. He was Professor at the National Nuclear Science and Technology Institute, and in 1995 he was appointed Research Director. The entire body of his work on the chemistry of actinides was recognised by the Academy of Science in 2005 with the Yvan Peyches Award.

Professor Madic gave a lot to science and was a great scientific leader in the development of minor actinide separation processes. For more than 15 years he also played a leading role in the building of a European community in partitioning. This community has grown with the years and with the successive integration of different projects, of different teams. There is now, thanks to his great contribution, a large network with numerous skills and tools spread all over Europe, joined today within the ACSEPT Project. Charles Madic deeply marked the scientific community, and in particular young scientists, by transmitting his passion for scientific research through numerous PhD theses and teaching courses.

The ACSEPT Project is dedicated to his memory.

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Development and demonstration of a new SANEX process for actinide(III)/lanthanide(III) separation using a mixture of CyMe$_4$BTBP and TODGA as a selective extractant

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Abstract

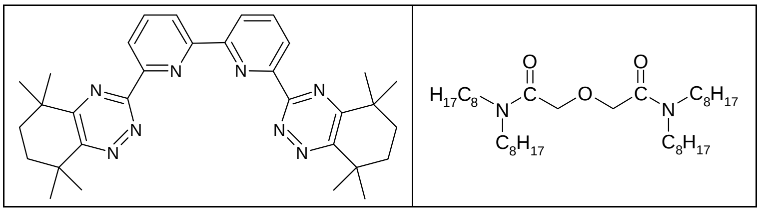
Within the framework of the European collaborative project ACSEPT, a new SANEX partitioning process was developed at Forschungszentrum Jülich for the separation of minor actinides (americium and curium) from lanthanide fission products in spent nuclear fuels. The development is based on previous and new batch solvent extraction studies, small single-centrifuge tests and on flow sheet design by computer code calculations. The mixture composed of 6,6´-bis(5,5,5,8,8-tetramethyl-5,6,7,8-tetrahydrobenzo-[1,2-4]trizazin-3-yl)°[2,2´]°bipyridine (CyMe$_4$BTBP) and N,N,N´,N´-tetra-octlyl-3-oxapentan-diamide (TODGA) dissolved in n-octanol was used as an efficient extractant. A spiked continuous counter-current test was carried out in miniature centrifugal contactors with the aid of a 20-stage flow sheet consisting of 12 extraction, 4 scrubbing and 4 stripping stages. With the aid of this flow sheet, more than 99.9% of the trivalent actinides Am(III), Cm(III) and Cf(III) were extracted and back-extracted. High feed decontamination factors over 1 000 were obtained for these actinides. The trivalent lanthanides were completely directed to the raffinate of the process. The actinide(III) product stream was only contaminated with less than 0.5 mass % of the initial lanthanides, such that the ambitious aim for a P&T scenario of < 5% was achieved.

Introduction

The selective partitioning of MA from fission products and separate treatment by transmutation can considerably improve long-term safety. Several processes have been developed for the co-extraction of americium and curium from the PUREX waste stream containing lanthanides and other fission products [1]. European research over the last decade, *i.e.* in the NEWPART, PARTNEW and the recent EUROPART programmes, has resulted in the development of a combined DIAMEX and SANEX process [2,3]. Both processes are based on the co-separation of trivalent actinides and lanthanides (*e.g.* by DIAMEX or TODGA/TBP process), followed by the subsequent actinide(III)/lanthanide(III) group separation in the SANEX process.

In the context of a European actinide partitioning strategy, the separation of trivalent actinides (americium and curium) from lanthanides is a key step but it still causes considerable problems due to the chemical similarity of the two groups of elements. This separation step can be achieved using soft donor nitrogen-bearing ligands, for instance 2,6-di(1,2,4-triazine-3-yl)pyridines (BTP). Counter-current SANEX tests with real highly-active feed solutions have shown promising results, but a major drawback is the low stability against hydrolysis and radiolysis [4]. The BTB molecules were further developed by the University of Reading in order to increase the chemical stability and ability to strip extracted metals. This was achieved by adding one more pyridine ring in the centre of BTP molecules, which produced the BTBP class ligands (see Figure 1). The $CyMe_4BTBP$ was introduced as a potential SANEX extractant for the separation of actinides(III) from lanthanides(III) [5]. Unfortunately, the extraction and stripping kinetics are rather slow and a phase transfer catalyst, such as a malonamide (DMDOHEMA), must be added for the development of a reversible extraction process [6].

Figure 1: $CyMe_4BTBP$ and TODGA mixture for the SANEX process



Within the framework of the process-oriented work at Forschungszentrum Jülich, an alternative process was developed using the diglycolamide TODGA as a phase transfer catalyst. Since TODGA itself is a very efficient extractant for the co-extraction of actinides(III) and lanthanides(III) [7], only small amounts of TODGA were used for the optimisation of the formulation of the SANEX process. The extractant was a mixture of 0.015 mol/L $CyMe_4BTBP$ + 0.005 mol/L TODGA in n-octanol as diluent. Batch extraction and single centrifuge tests were carried out, and the results were used to design a flow sheet by computer code development. The new SANEX process for actinide(III)/lanthanide(III) separation was tested in miniature centrifugal contactors with the aid of a 20-stage flow sheet consisting of 12 extraction, 4 scrubbing and 4 stripping stages. In the present paper, the results of the demonstration of the SANEX process will be presented and discussed.

Experimental

$CyMe_4BTBP$ was synthesised and provided by Reading University, UK. Details of synthesis, stability and general extraction properties can be found in [6]. TODGA was synthesised in Jülich according to a known synthesis route based on diglycolyl chloride and dioctylamine as starting materials [8]. The extractant mixture was prepared by dissolving 0.015 mol/L $CyMe_4BTBP$ and 0.005 mol/L TODGA in n-octanol as diluent. The An(III)/Ln(III) feed was an ^{241}Am, ^{244}Cm, ^{252}Cf, ^{152}Eu tracered synthetic solution whose lanthanide concentration corresponded to a real TODGA/TBP re-extraction process solution [9,10]. The composition is shown in Table 1. An optimised 20-stage flow sheet (Figure 2) was designed by a computer code [11] using data from batch and single centrifuge experiments [12].

Table 1: Composition of the synthetic An(III)/Ln(III) feed used for the SANEX process

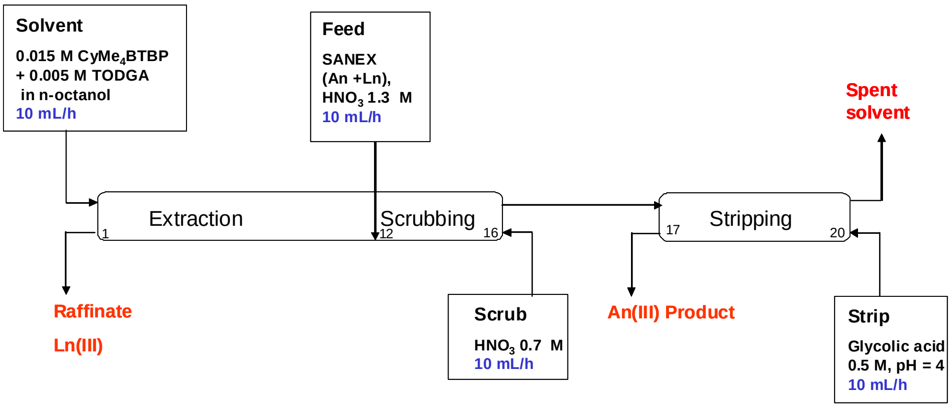
Element	Concentration (mg/L)	Nuclide/activity (MBq/L)
Am	Traces	^{241}Am/2.5
Cm	Traces	^{244}Cm/2.4
Cf	Traces	^{252}Cf/1.6
Ce	383	
Eu	129	^{152}Eu/3.3
Gd	85	
La	205	
Nd	716	
Pr	192	
Sm	142	
Y	55	

The SANEX test was carried out using miniature centrifugal extractors in a counter-current mode. Details of the experimental set-up can be found in [8]. The pumps used in the test were piston pumps, which were able to produce small flows controlled by weighing. Since only 16 stages were available, stripping was performed in a separate test with 4 stages after cleaning the apparatus. The solvent loaded with actinides(III) (coming from stage 16) was collected in 2 batches for this purpose and the fraction collected after the attainment of the steady state was used for stripping. The continuous process described in the following consists of three process sections:

- Twelve (12) extraction stages for the extraction of Am(III), Cm(III) and Cf(III) from 1.3 mol/L HNO_3.
- Four (4) scrubbing stages for back extraction of the poorly co-extracted lanthanides with 0.7 mol/L HNO_3.
- Four (4) stripping stages for the back extraction of the trivalent actinides with 0.5 mol/L glycolic acid. The stripping of the extracted trivalent actinides is also thermodynamically possible with diluted nitric acid, but batch experiments show that the kinetic is rather slow. Therefore the hydrophilic complexing agent, glycolic acid (solution set to pH 4) was used for stripping [6].

No hydrodynamic problems were encountered during the SANEX test. All of the phases collected were clear and free of entrainment. After the test, the following analyses were carried out on all stages (aqueous and organic) and all collected samples, including the samples to determine the transient state: γ spectroscopy for ^{241}Am, ^{152}Eu, α spectroscopy for ^{241}Am, ^{244}Cm, ^{252}Cf and ICP-MS for all inactive lanthanides(III) elements. The acidity profile was determined for the aqueous phase by titration with NaOH.

Figure 2: Flow sheet of the SANEX process using $CyMe_4BTBP$ + TODGA as extractant



Results and discussion

The most important results of gamma, alpha and ICP-MS measurements are shown in Table 2. The process decontamination factors $DF_{Am/Ln}$ and $DF_{feed/raff}$ were calculated according to the following Eqs. (1) and (2), where Q is the flow rate in mL/h and C is the concentration of the element in mg/L:

$$DF_{Am/Ln} = \frac{Q_{feed} \cdot C_{Ln\ \ feed} \cdot C_{Am\ organic\ \ product\,(stage16)}}{Q_{organic\ \ phase} \cdot C_{Am\ \ feed} \cdot C_{Ln\ organic\,product\,(stage16)}} \tag{1}$$

$$DF_{feed/raff} = \frac{Q_{feed} \cdot C_{feed}}{Q_{raff} \cdot C_{raff}} \tag{2}$$

The main conclusions drawn from Table 2 can be summarised as follows. The trivalent actinides americium and californium were quantitatively extracted (> 99.9%) and completely recovered after stripping with glycolic acid. High decontamination factors over 1 000 between the feed and the raffinate were achieved for these actinides. Curium was also extracted up to > 99.7%, but 0.3% still remained in the raffinate, so a lower DF of 314 was achieved here. The results of the back extraction show that four stages were sufficient for the actinide (III) stripping as also shown in Figure 3. However, gamma spectrometric analysis shows that 0.04% of ^{241}Am was still found in the spent solvent. It can be assumed that the behaviour of Cm and Cf is similar, as the background level of alpha spectrometry was slightly higher than the gamma measurement (Figure 3). The mass balances were good for the actinides(III). For lanthanides, however, an excess in the raffinate was observed. High decontamination factors were measured between americium, here as a representative of actinide (III) and the lighter lanthanides La, Ce, Pr and Nd. This was in agreement with the results obtained in batch experiments. As shown in Table 2 and Figure 4, acid scrubbing efficiently reduces the co-extraction of the lighter lanthanides. Since the major rare earth elements are usually the lightest ones in genuine irradiated fuels, this is a very positive result. The higher lanthanides Sm, Eu and Gd, which are much less concentrated in the feed solution, are transported to the organic phase to a small extent and efficiently back-extracted together with the actinides. The co-extraction of Gd (1.6%,), Eu (0.8%) and 3.2% of Y can be further reduced by introducing more scrubbing stages. If we consider the total lanthanide inventory, the actinide(III) product stream was contaminated with less than 0.5 mass % of the initial lanthanides. This is a very good result in view of the P&T strategy for minor actinides.

Table 2: Main results of the SANEX process

Elements	% in raffinate	% in An(III) prod. (17 aq)	% in solvent (stage 20)	DF feed/raff	DF Am/Ln
Ce	121	0.11	0.0044	–	6 184
Eu	112	0.76	0.0018	–	102
Gd	111	1.62	0.0010	–	56
La	121	0.10	0.0015	–	41 449
Nd	120	0.10	0.0007	–	14 746
Pr	121	0.11	0.0013	–	11 802
Sm	116	0.23	0.0010	–	559
Y	110	3.22	0.0054	–	25
^{241}Am (γ)	0.08	99.9	0.038	1 161	
^{152}Eu (γ)	105	0.8	0.022	0.95	
^{244}Cm (α)	0.32	99.2	d.l.	314	
^{252}Cf (α)	d.l.	107.6	d.l.	> 1 000	
^{241}Am (α)	d.l.	99.2	d.l.	>1 000	

Figure 3: Experimental organic and aqueous Am(III), Cm(III) and Cf(III) concentration profiles during the SANEX test

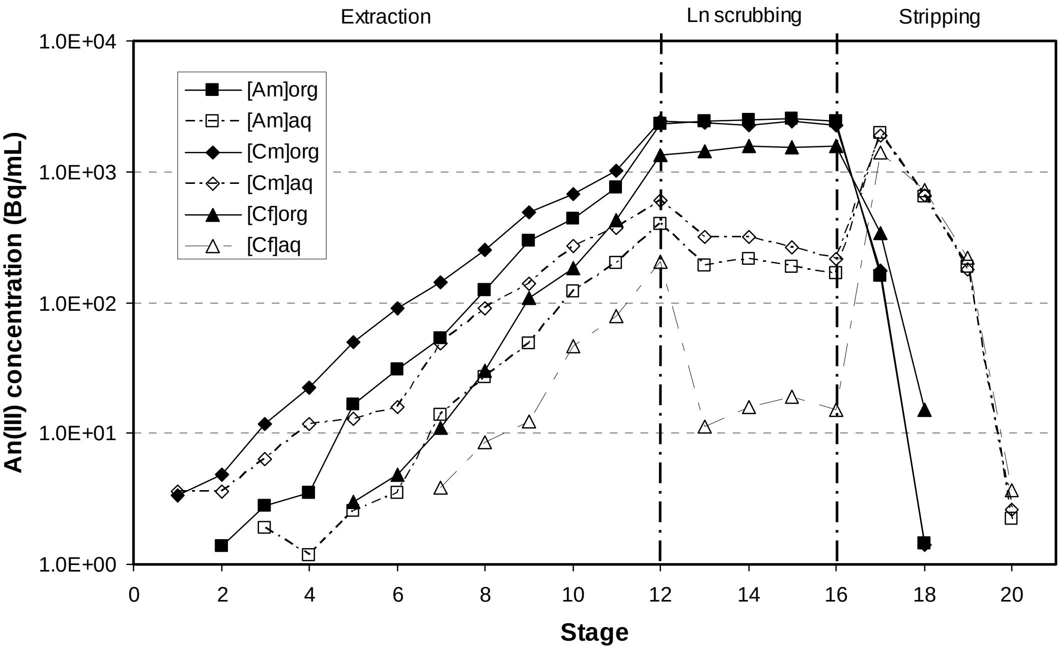


Figure 4: Experimental organic and aqueous Am(III) and Eu(III) concentration profiles during the SANEX test

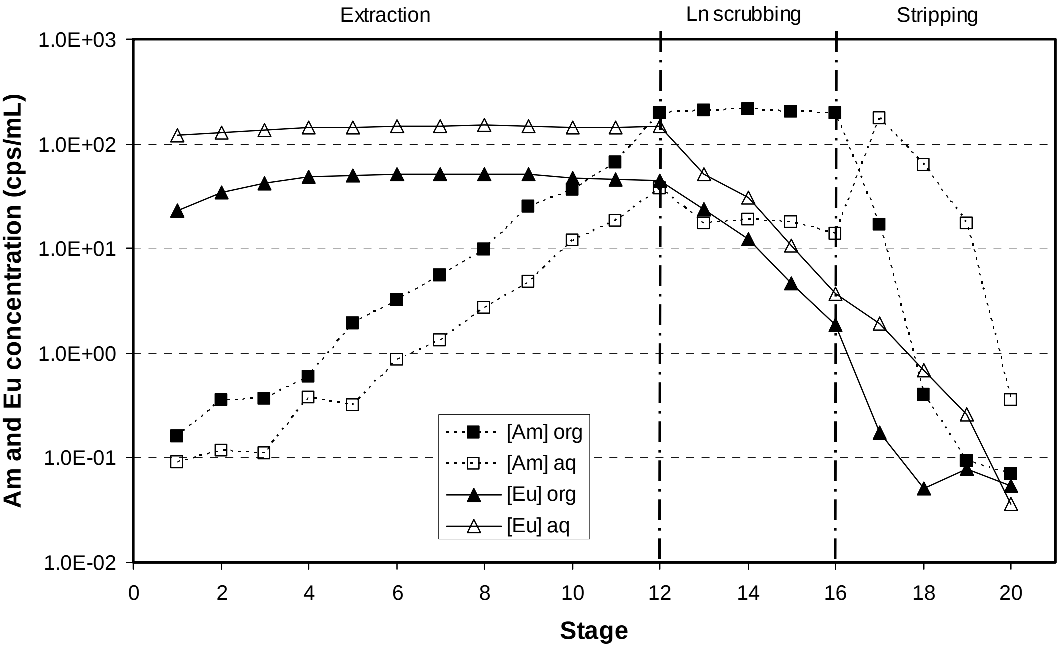
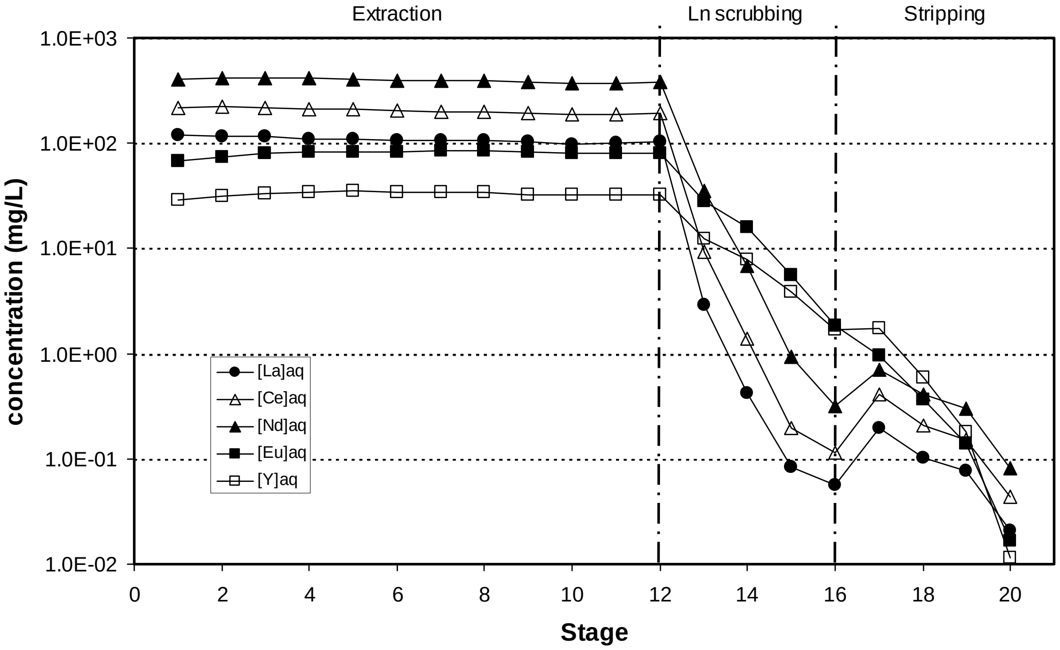


Figure 5: Experimental aqueous concentration profiles of some lanthanides during the SANEX test



Conclusions

The results of the spiked counter-current centrifugal contactor test showed that the mixture of CyMe$_4$BTBP and TODGA in n-octanol as diluent is a promising solvent for the selective separation of trivalent actinides over lanthanides(III) from a rather acidic solution of 1.3 mol/L HNO$_3$. This was demonstrated in an optimised flow sheet involving 20 stages. With the aid of this flow sheet, more than 99.9% of the trivalent actinides were extracted and back-extracted. The actinide(III) product stream was only contaminated with less than 0.5 mass % of the initial lanthanides, such that the ambitious aim of < 5% was achieved. Considering the slow extraction kinetics and the efficiency of the extraction stages, the experimental results are in good agreement with the prediction of the calculations.

Acknowledgements

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Recent progress on R&D of innovative extractants and adsorbents for partitioning of minor actinides at JAEA

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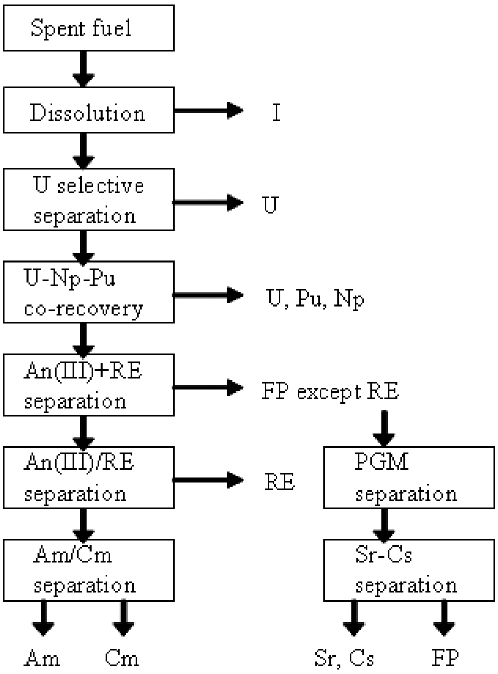
Abstract

The R&D effort on partitioning of minor actinides (MA) at the Japan Atomic Energy Agency (JAEA) has been concentrated on development and improvement of innovative extractants and adsorbents as the fundamental studies and of MA recovery process as the advanced aqueous reprocessing system in fast reactor cycle technology development (FaCT) project. This paper reviews current status and prospects of the R&D activities on the partitioning of MA at JAEA.

Introduction

Long-lived minor actinides MA (Np, Am and Cm), heat generating elements Sr-Cs, long-lived fission products (LLFP) such as Tc, platinum group metals (PGM) and rare metals in spent nuclear fuel will be target elements for partitioning and transmutation (P&T) system. The partitioning process for the elements above can be constructed by combining several separation steps, together with the reprocessing process of major components of the spent fuel, U and Pu. Figure 1 shows one of the concepts for the separation process flow. The reprocessing process of U and Pu is strongly dependent on the type of the spent fuel and the product of U or Pu. Neptunium, one of MA, can also be separated with U and Pu. After the separation of U and Pu, the partitioning of MA from FP starts in this process flow with two separation steps, *i.e.* the first step is the recovery of trivalent actinides (An(III)) with rare earth elements (RE) and the second step the separation of An(III) from RE, where RE contains Y and lanthanides in the spent fuel.

Figure 1: Separation process flow



The Japan Atomic Energy Agency (JAEA) has been investigating the advanced aqueous reprocessing system known as the New Extraction System for TRU Recovery (NEXT) process in fast reactor cycle technology development (FaCT) project [1], together with various partitioning methods as alternative technologies by using solvent extraction and chromatography. Those are summarised in Table 1.

This paper reviews some of the R&D activities on aqueous partitioning of MA at JAEA.

Recovery of An(III) with RE by solvent extraction

TRUEX

The TRansUranium Extraction (TRUEX) process is a component of UREX+, and recovers An(III) and RE from highly active waste solutions. Application to the raffinate generated from reprocessing FBR spent fuel was studied. CMPO(n-octyl(phenyl)-N,N-diisobutyl carbamoylmethylenephosphine oxide) strongly co-ordinates to An(IV) and An(VI) ions, and U(VI) and Pu(IV) is hardly stripped with a dilute nitric acid solution. By utilising a founding that an alkaline scrub is effective to strip their ions, the flow sheet which uses “salt-free” reagents free from metallic cations was prepared in order to reduce the amount of secondary wastes. A counter-current experiment using a raffinate from processing FR JOYO irradiated fuel was conducted [2]. The fraction of Pu that retained in the used solvent was suppressed to 0.5%.

Table 1: Recent activities on aqueous partitioning at JAEA

Separation step	NEXT process	Other methods & technologies
U selective separation	Crystallisation	• Precipitation by pyrrolidone • Extraction by TBP (modified PUREX) • Extraction by N,N-dialkylamide
U-Np-Pu co-recovery	Co-extraction by TBP	• Extraction by N,N-dialkylamide
An(III)+RE separation	**Extraction chromatography (CMPO, TODGA)**	**• TRUEX (Extraction by CMPO)** **• Extraction by diglycolamide (TODGA)** • Extraction by CMPO with fluorinated diluent • Ion exchange (tertiary pyridine resin) • Extraction by DIDPA
An(III)/RE separation	**Extraction chromatography (BTP, HDEHP)**	**• SETFICS (extraction by CMPO with DTPA)** **• TALSPEAK (extraction by DIDPA with DTPA)** **• Extraction by TPEN, TPA, PDA, BTP, etc.** • Extraction chromatography (PDA) **• Ion exchange (tertiary pyridine resin – HCl-MeOH)**
Am/Cm separation	–	**• Ion exchange (tertiary pyridine resin – HNO_3-MeOH)** • Extraction chromatography (future work)
Sr-Cs separation	–	• Inorganic ion exchanger (zeolite, titanic acid) • New mixed oxide adsorbent • Extraction chromatography • Hybrid adsorbent (micro-capsule)
PGM separation	–	• Electrolytic extraction • Precipitation by denitration

DGA extraction

Phosphorus-free extractants consisting of carbon, hydrogen, oxygen and nitrogen (CHON principle) have been investigated to reduce secondary solid wastes generated from spent organophosphorus extractants such as CMPO, HDEHP[di(2-ethylhexyl) phosphoric acid], and DIDPA (diisodecylphosphoric acid). During the course of the studies on the modification of a bidentate malonamide, we developed diglicolamide (DGA) compounds for the recovery of long-lived actinide ions. TODGA (N,N,N′,N′-tetraoctyldiglycolamide), one of the DGA compounds, has several advantages, *i.e.* high distribution coefficients for An(III) and An(IV) from concentrated nitric acid to n-dodecane, high solubility in aliphatic diluents, high stability for hydrolysis and radiation [3]. Further, the lipophilic and hydrophilic properties of DGA compounds can be changed by modifying its alkyl groups attached to amidic nitrogen atoms [4].

We found that DGA compounds with longer alkyl groups, *i.e.* TDDGA (N,N,N′,N′-tetradecyl-diglycolamide) and TDdDGA (N,N,N′,N′-tetradodecyl-diglycolamide) have higher extraction capacities for divalent, trivalent and tetravalent metal ions than those of TODGA [5]. As a demonstration of recovery of trivalent ions from fission products, counter-current extraction and separation of Nd from Sr, Zr and Pd by TDdDGA was carried out, where Nd is used as a representative ion of An(III) and RE and Sr, Zr and Pd as typical fission products. On the basis of a simple simulation calculation using the distribution ratios obtained by batch experiments, 0.1 M TDdDGA/n-dodecane and 1 M HNO_3 + 0.2 M H_2O_2 + 0.02 M Na_2 EDTA were selected for the extraction solvent and for the feed and scrub solutions, respectively. The results showed that more than 99.94% of Nd was recovered and separated from the other metal ions [6].

Separation of An(III) from RE by solvent extraction

SETFICS

The TRUEX process was improved to crudely separate RE from An(III) and named as the SETFICS (Solvent Extraction for Trivalent f-element Intra-group Separation in CMPO-complexant System) process [7]. A polyaminoacetic acid DTPA (diethylenetriaminepentaacetic acid) was employed to obtain selectivity on An(III)/RE and a flow sheet was established to selectively strip An(III). After nitric acid is rejected from a loaded solvent that contains An(III) and Ln(III), An(III) is stripped with a

DTPA-nitrate solution. A counter-current experiment with centrifugal contactors was conducted and obtained the loss of Am and Cm of 0.16% and 0.33-0.39%, respectively [8]. The decontamination factors for La, Ce, Pr, Nd and Sm were > 3, 100, > 10, 1.8 and 1.9, respectively.

In order to attain a higher metal loading on a CMPO mixed solvent, a fluorine compound fluoropol-732 as a diluent was examined [9]. Am of > 99.97% was recovered, and contamination with light lanthanides was suppressed as 0.5%, 0.04%, 0.3% and 4.4% for La, Ce, Pr and Nd, respectively.

TALSPEAK

The separation method of An(III) from RE by using HDEHP as extractant and DTPA as a complexing agent is called TALSPEAK (Trivalent Actinide-lanthanide Separation by Phosphorous Reagent Extraction from Aqueous Complexes). This method is also one of the components of UREX+.

DIDPA instead of HDEHP was used for MA separation step in the four-group partitioning process developed at the former JAERI (Japan Atomic Energy Research Institute), presently JAEA. We examined a selective stripping method by DTPA for Am(III) and Cm(III) from RE in DIDPA solvent. The performance of the selective stripping and the optimisation of the separation process were examined by a simulation code which uses the values of distribution ratios of Am(III) and RE by batch experiments. The validity of the simulation code was confirmed by comparing the results of the simulation with the results of continuous stripping tests of RE with DTPA solution from the DIDPA solvent using mini-mixer-settlers [10].

New extractants

One promising approach for separating An(III) from RE is to use soft-donor ligand based on its preferable co-ordination to softer An(III). Nitrogen-containing heterocycle-based multidentate ligands have been presently attracted by their combustibility (CHON principle).

We reported that some separation systems using TPEN(N,N,N′,N′-tetrakis(2-methylpyridyl)-ethylenediamine) [11] or TPA (tris(2-pyridylmethyl)amine) have high separation factors of An(III)/RE, $SF_{(An/RE)}$. Furthermore, we designed two ligand systems which were expected to efficiently separate An(III) from RE. One is an "Oligo-pyridine" ligand containing multiple pyridines [12] and another is a "Chirality-controlled" ligand containing chiral centers in the ligand [13]. We have synthesised more than thirty ligands and found out the ability on the separation of An(III) from RE, with $SF_{(Am/Eu)} > 10$ for many ligands.

We have developed new ligands which have sufficiently high stability for hydrolysis and radiolysis, high lipophilicity, and sufficient extractability and selectivity in higher concentration of nitric acid. On the basis of the results on the extraction of An(III) and RE by DMDPhPDA (N,N′-dimethyl-N,N′-diphenylpyridine-2,6-dicarboxyamide) which extracts An(III) more selectively than RE from HNO_3 solution [14], we have investigated some pyridine dicarboxyamide (PDA) compounds by modifying its alkyl groups attached to amidic nitrogen atoms. N,N′-dioctyl-N,N′-diphenylpyridine-2,6-dicarboxyamide, one of PDA, was newly synthesised and applied to extraction chromatography as an application. PDA was successfully impregnated into Amberlite XAD-4 resin. The separation of Am(III) from Eu(III) in 5 M HNO_3 was sufficiently achieved by using PDA/XAD-4 column (5 mm $\phi \times 20$ cm) [15].

Application of chromatographic technology

Extraction chromatography

Solvent extraction has been studied on An(III) separation from a highly acidic raffinate generated in spent fuel reprocessing. Recent research is focused on direct and selective extraction of An(III) cations as well as An(III) separation from Ln(III). The chemical structure of extractants currently investigated became more complex, *e.g.* a nitrogen-donor ligand TPTZ (2,4,6-tris-(2-pyridyl)-1,3,5-triazine) was replaced by BTP (bis-triazinyl-pyridine). A compound of a large molecular weight often brings issues on solubility into a diluent, kinetics, hydrolysis and radiolysis. In consequence, a recovery system of An(III) by solvent extraction would be a large size because of a combined process that uses a couple of extractant, use of an extractant that should be highly diluted. For the countermeasure, development on application of extraction chromatography was initiated.

Since highly active raffinate (HAR) containing An(III) induces decomposition of organic compounds, a rapid adsorption and elution are favored. A porous silica particle as a support is effective because of its rigid and uniform structure. An extractant is retained on a silica support coated with styrene-divinylbenzene for use, thus, separation chemistry is similar to that of solvent extraction. A flow sheet study is underway to select promising extractants as the state of the art.

A device of extraction chromatography needs an extensive development for its realisation including the subjects of safety such as solid particles contained in an HAR, radiolysis from alpha- and gamma-rays and safety design, the subjects of devices that confines highly active material and remote handling, and the subjects of control that requires batch-wise operation.

As a scenario concerning to safety, a case in which An(III) is retained on the adsorbent and hardly eluted shall be considered. An adsorbent is a composite of silica and organics compounds, thus, is decomposed under a higher temperature. A higher limiting value on thermal reaction can be set compared with solvent extraction, since extraction chromatography is free from a diluent which limits the temperature of operation. In order to investigate the limiting temperature, TG/DTA analysis was conducted for CMPO or HDEHP/SiO_2-P adsorbent [16]. Both extractant decomposes at about 220°C.

Tertiary pyridine resin

Chromatographic separation method has been investigated to realise a novel, multi-functional reprocessing technology. Tertiary pyridine resin with two functions of ion exchanger and soft-donor was dedicated to the experiments. Two important facts were found in the past basic experiments that MA(III)/Ln(III) separation was possible in HCl media, and Am(III)/Cm(III) mutual separation was enhanced under the methanolic HNO_3 media. Based on this knowledge, highly irradiated (143.9 GWd/t) mixed-oxide fuel from the experimental FR JOYO was dedicated to the hot tracer flow sheet tests. As a result of a series of tests, selective sorption of ^{106}Ru (PtG) with ^{125}Sb by dilute HCl (0.5 M) medium, and extensive group separation of MA, Ln and Pu was confirmed by HCl (12 M, 0.1 M) media. Successively, mutual MA separation was obtained in methanolic (60%) conc. HNO_3 medium. With a three-step separation, highly pure Am and Cm products were individually obtained. The decontamination factor (DF) of ^{137}Cs and Ln (^{155}Eu, ^{144}Ce) against the Am product exceeded 3.9×10^4 and 1.0×10^5, respectively. The DF of ^{243}Cm against the Am product was larger than 2.2×10^3. The contents of ^{137}Cs, Ln and ^{243}Cm in Am product were very low, less than 2 ppm [17].

Presently, development of anti corrosive materials such as Ta and Hastteloy-B (Ni-28Mo), etc., for high-temperature, concentrated HCl media, thermochemistry of tertiary pyridine resin under the methanolic HNO_3 media are mainly under the investigation in the reference with those of HNO_3-based PUREX environment [18].

Conclusions

Solvent extraction and extraction chromatography have mainly been investigated for the partitioning of MA and the selection of the extractant is an essential point to achieve the selective separation of An(III) in both cases. A large number of compounds as the extractants of An(III) have been synthesised and tested, but the definitive one is not yet been determined from the viewpoint of the engineering level on separation, stability and safety. Therefore, the efforts to develop and improve the innovative extractants should be continued to achieve the partitioning process of MA, especially for the separation of An(III) from RE.

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Fission product partitioning and integrated waste management – advanced approaches and opportunities

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Abstract

Nuclear energy will be a critical resource for the future, especially as countries seek energy independence, meet rising energy demand, and address the challenges of global climate change. Nuclear energy must play a major role and there needs to be not only status quo, but a large increase in its contribution to overall energy production (the number of reactors required is large). Significant challenges to this sustainable future for nuclear energy include waste management. The essential, sustainable future for nuclear energy warrants an integrated, comprehensive approach to the fuel cycle and nuclear waste management.

Advanced separations integrated with waste form production and performance provides opportunities for flexibility and ultimately fuel cycle system optimisation. Removal of uranium and transuranics and management of fission product storage and waste forms as part of a closed fuel cycle would impact the potential technical capacity of a geologic repository. Many factors potentially control the technical capacity of a geologic repository including: i) post-closure repository performance as measured by potential radiation dose; ii) physical area available with suitable geologic and hydrologic characteristics; iii) waste form volume; iv) waste thermal output. These factors must be evaluated as part of a fuel cycle system. For example, fission products contribute to the heat load of a geologic repository, with the significance and resultant challenges dependent on the geologic setting and safety case for a given repository. Therefore, many options exist for fission product heat management involving various combinations of strategies including spent nuclear fuel storage prior to reprocessing, waste form decay storage after reprocessing, and dedicated disposition paths.

Overall, if the repository capacity benefits from closing of the fuel cycle are realised, this will reduce the amount of materials destined for the repository and also could eventually allow for an evolution in the design of repositories, which could lead potentially to significant cost savings and system efficiencies. A closed fuel cycle also provides opportunities to develop advanced waste forms that are more robust than spent fuel and tailored to the specific geochemical environments of a repository. Work remains to be done to develop and successfully demonstrate the advanced fuel cycle technologies and also to better understand the potential increases of the technical capacity of the repository as part of a closed fuel cycle. Additional work includes development of integrated waste and fission product management strategies, further technical analyses of constraints on repository capacity and development of potential advanced waste forms.

Introduction

The US Global Nuclear Energy Partnership/Advanced Fuel Cycle Initiative (GNEP/AFCI) programme aims to recycle nuclear fuel using advanced safe and secure technologies to recover more energy and reduce the amount of high-level waste (HLW) that must be managed and disposed. Several of the GNEP/AFCI goals pertain directly to waste management and disposition:

- Reduce the environmental and financial burden and uncertainty associated with long-term nuclear waste management.
- Optimise nuclear waste management by:
  - minimising the risk of waste that needs to be handled or stored;
  - producing only solid wastes in robust waste forms;
  - recycling and reusing materials to the maximum extent possible.
- Support the near-term deployment of fuel cycle technologies (20 years) as well as define longer-term deployments of next-generation technologies (50 years).
- Make the closed fuel cycle as economical as possible.
- Reduce the number of required US geologic waste repositories needed for the remainder of this century.

This paper discusses the reasons for and importance of fission product (and minor actinide) partitioning and the benefits that could be achieved in regard to the management and disposition of wastes that would be generated by an advanced nuclear fuel cycle. It also presents activities that are being conducted in the GNEP/AFCI programme, striving to meet the goals presented above.

Fission product partitioning

Current reprocessing techniques, such as PUREX, recover uranium and plutonium from used nuclear fuel for subsequent use as fuel in nuclear reactors. Advanced reprocessing techniques, such as the UREX+ suite of extractions [1], recover minor actinides as well as uranium and plutonium. The GNEP/AFCI programme plans to include the minor actinides in transmutation fuels or targets for irradiation in fast nuclear reactors, with the goal of effectively removing most of the minor actinides from the waste.

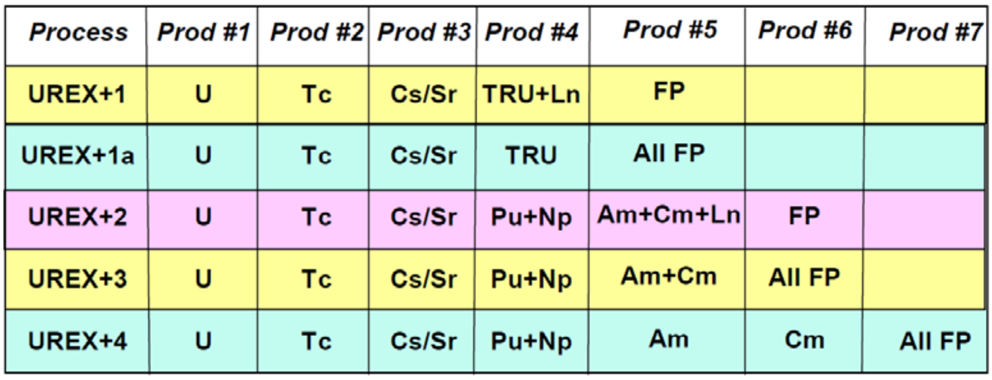
The waste streams that result from current reprocessing techniques including both fission products and minor actinides, are high-level nuclear waste (HLW), and require isolation in a deep geologic repository. Advanced reprocessing techniques being developed under the GNEP/AFCI allow for minor actinide transmutation and the possibility to further partition the fission products, potentially leading to benefits in managing and disposing the remaining wastes.

The US GNEP/AFCI programme has been developing a suite of processes known collectively as UREX+, shown in Figure 1 [1]. Each of the UREX+ processes involves the partitioning of technetium, caesium and strontium. The UREX+ process concepts differ in how they partition the transuranic elements (TRU) and remaining fission products (*e.g.* lanthanides and transition metals). Head-end steps (chopping and voloxidation) and dissolution result in the release of volatile elements (carbon, iodine, krypton, xenon, tritium) for subsequent capture. Undissolved solids, containing fission products and some actinides, residuals from the dissolution step must also be recovered for disposal. The UREX +1a process, shown schematically in Figure 2, is the reference process in the US AFCI/GNEP programme [1].

The waste streams from the UREX +1a process and the reference waste forms being considered in the GNEP/AFCI programme are shown in Figure 3. It can be seen that a variety of waste forms will be generated that must be managed and disposed. Volatile elements (iodine, krypton, carbon, tritium) will be captured out of the off gas and immobilised. The separation of Cs and Sr allows for their management in a long-term (~100 year) storage facility where ^{134}Cs, ^{137}Cs and ^{90}Sr can decay, reducing their decay heat prior to disposal. The two major waste forms are metal alloy and glass. Some residual TRU is expected to report to the lanthanide fission product waste stream, where they it would be isolated in glass and to the undissolved solids where it would be isolated in a metallic alloy.

Figure 1: The suite of UREX+ processes [1]

FP – fission products (balance); LN – lanthanide fission products only



Process	*Prod #1*	*Prod #2*	*Prod #3*	*Prod #4*	*Prod #5*	*Prod #6*	*Prod #7*
UREX+1	U	Tc	Cs/Sr	TRU+Ln	FP		
UREX+1a	U	Tc	Cs/Sr	TRU	All FP		
UREX+2	U	Tc	Cs/Sr	Pu+Np	Am+Cm+Ln	FP	
UREX+3	U	Tc	Cs/Sr	Pu+Np	Am+Cm	All FP	
UREX+4	U	Tc	Cs/Sr	Pu+Np	Am	Cm	All FP

Figure 2: UREX +1a process [1]

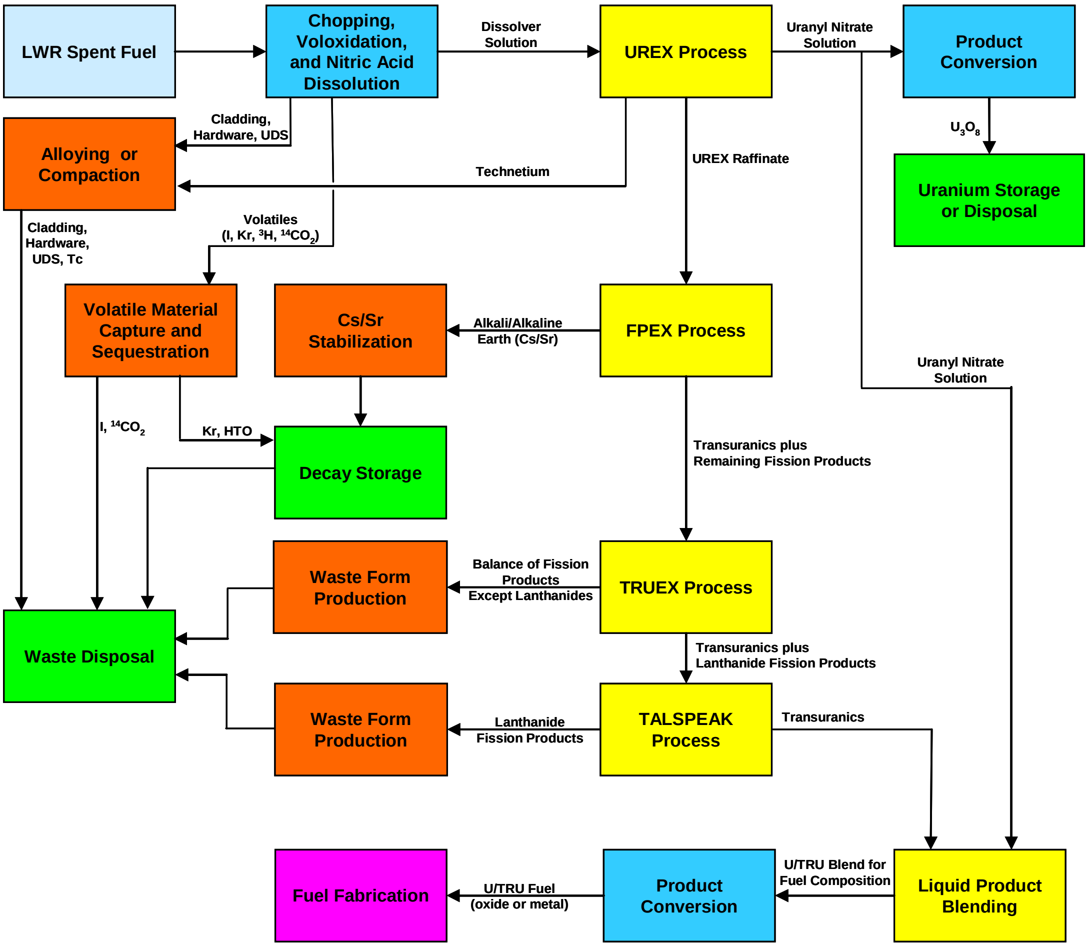
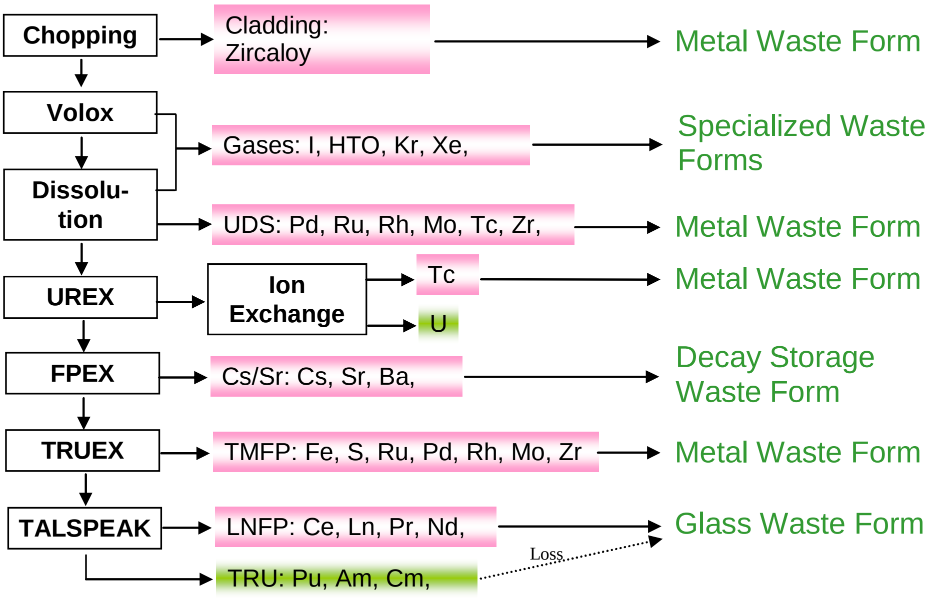


Figure 3: UREX +1a reference waste streams and waste forms

Product materials are highlighted in green

HTO – tritiated water, UDS – un-dissolved solids, TMFP – transition metal fission products, LNFP – lanthanide fission products, TRU – transuranics



Benefits of fission product partitioning

The potential benefits, in terms of managing and disposing nuclear waste, that could result from the removal of minor actinides and fission product radionuclides (^{137}Cs and ^{90}Sr) from the waste stream have been evaluated [2-5]. These potential benefits include:

- reduction in the volume of HLW that must be disposed in a deep geologic disposal facility as compared to the direct disposal of spent nuclear fuel;
- reduction in the amount of long-lived radioactive material (*e.g.* minor actinides) that must be isolated in a geologic disposal facility (reduction of source term);
- reduction in decay heat allowing for increased thermal management flexibility, potentially increasing emplacement density.

Volume reduction

The volume-to-mass ratio of commercial spent nuclear fuel is approximately 0.46 m^3/MTHM [5]. The volume of each of the reference UREX +1a waste streams that would be generated from the processing of commercial spent nuclear fuel has been estimated and is shown in Table 1 [6]. These waste streams would require management and ultimate disposal. Note that the volume of krypton is not included in Table 1 because it is assumed that it could be released after a period of decay storage (^{85}Kr has a 10.7 year half-life). Tritium will likely be disposable as low-level radioactive waste and is also not included in Table 1. A comparison of the total volume of these waste streams with the volume of commercial spent nuclear fuel is also shown in Table 1. It must be recognised that Table 1 presents waste form volumes exclusive of packaging and how the waste is further isolated in canisters and/or waste packages and how the canisters and/or waste packages are emplaced in a disposal facility would influence the actual disposal volumes and the corresponding volume reduction factor.

Table 1 shows that the overall volume of all the UREX +1a reference waste forms will be smaller than the volume of spent nuclear fuel from which the waste derived. Note that the volume of low-level radioactive waste that would be generated from UREX +1a processing are not shown in Table 1, but

Table 1: Estimated volume of UREX +1a reference waste forms [6]

Waste form	Potential waste form	Volume (m^3/MTHM)		Volume Fraction	
		Low	High	Low	High
I	Encapsulated zeolite	2.3E-03	9.3E-03	2.7%	5.1%
C	Grouted $CaCO_3$	1.9E-02	3.8E-02	22.4%	20.8%
Cs/Sr	Glass/ceramic	4.0E-03	2.7E-02	4.7%	14.8%
Tc/UDS/FP	Metal alloy	6.2E-03	1.8E-02	7.3%	9.9%
Ln	Glass	6.6E-03	1.8E-02	7.8%	9.9%
Hulls	Metal	3.8E-02	5.9E-02	44.8%	32.3%
Hardware	Metal	8.6E-03	1.3E-02	10.1%	7.1%
Total		**0.08**	**0.18**		
Volume reduction relative to SNF		**5.4**	**2.5**		

could be significant and will be higher than from the current once-through fuel cycle. The grouted $CaCO_3$, cladding hulls, and hardware wastes represent a significant fraction of the total volume (~60-75%). It may be possible to dispose of these wastes in a facility other than a deep geologic repository facility, further reducing the volume of material that would have to be disposed in such a facility.

While Table 1 shows that the volume of waste requiring disposal in a geologic repository that may be generated under an advanced nuclear fuel cycle may be significantly smaller than directly disposed spent nuclear fuel, secondary wastes would be generated in the fuel and waste processing steps that would also have to be disposed. The volume of these wastes may be significant. The draft Global Nuclear Energy Partnership Programmatic Environmental Impact Statement shows that the total the volume of waste that would require disposal (both as low- and high-level nuclear waste) is significantly larger under a closed advanced nuclear fuel cycle as compared to a once-through nuclear fuel cycle [7].

Source term reduction

Past analyses have shown that removing the TRU from the wastes that would be disposed in a deep geologic repository would result in a significant reduction in the long-term risk as compared to the direct disposal of spent nuclear fuel in a repository located in an unsaturated tuff formation (*i.e.* Yucca Mountain, Nevada) [4]. More current analyses of the performance of a disposal system in unsaturated tuff indicate that while TRU radionuclides do contribute to long-term disposal system risk, the risk is equally dominated by fission products (primarily ^{99}Tc and ^{129}I) as shown in Figure 4 [8].

The Nuclear Energy Agency led a study of how advanced nuclear fuel cycles would affect radioactive waste management [2]. The results of long-term assessments of disposal system risk indicated that ^{129}I, ^{79}Se, ^{126}Sn and ^{135}Cs, and ^{99}Tc are the dominant radionuclides in clay, granite, and salt media disposal environments. Minor actinides were found to be unimportant because of their low solubility in reducing environments.

Thus, while removing minor actinides from the waste would remove a source of hazardous material, the remaining fission products would still contribute significantly to the total risk and would require effective isolation. The reduction in the source term due to the removal of the minor actinides may allow for re-design of the engineered barrier system within a geologic repository while still protecting the public and workers from exposure to radiation. In addition, the partitioning of fission products, such as is envisioned in the UREX +1a process, could allow for their immobilisation in waste forms that match the chemistry of the particular fission products. This could result in improved waste form performance within a geologic environment and a further reduction in long-term risk.

Thermal management flexibility

The thermal density from decay heat of commercial spent nuclear fuel as packaged and placed in a repository is dominated by the heat generated mainly by the decay of fission products for the first 60 years and from actinide elements thereafter, as shown in Figure 5 [3]. The current design of the

Figure 4: Results of a total system performance assessment of a repository at Yucca Mountain, Nevada [8]

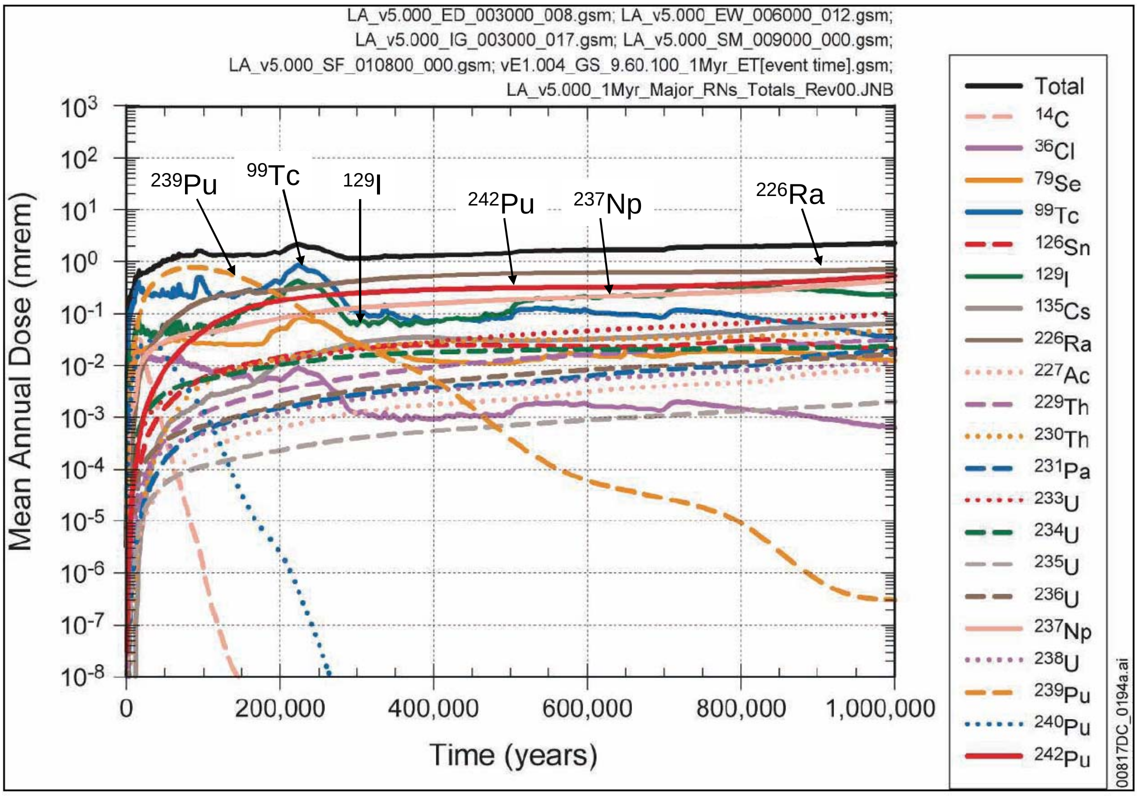
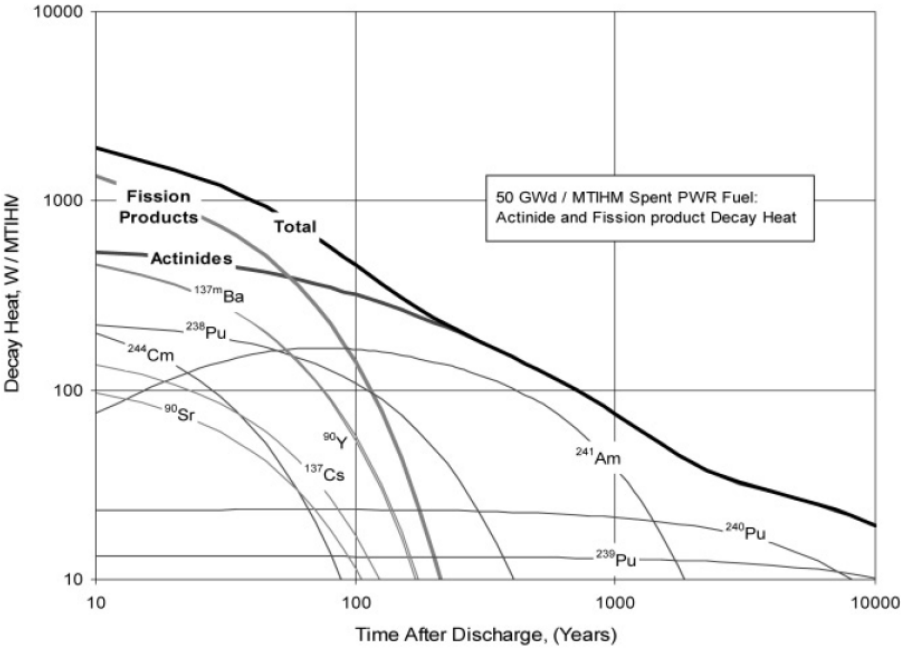


Figure 5: Dominant decay heat contributors in spent PWR fuel irradiated to 50 GWd/MTIHM [3]



sub-surface facilities at a proposed repository at Yucca Mountain is driven by the thermal density and has been chosen to satisfy thermal criteria related to the overall long-term performance of the repository (thermal aspects of design are not regulatory requirements). These criteria are [9]:

- maximum centre temperature between emplacement drifts of 96°C to ensure that the pillars between the emplacement drifts are free draining;
- maximum cladding temperature of 350°C
- maximum waste package surface temperature of 300°C;
- emplacement drift wall temperature below 96°C during the pre-closure period so as not to preclude cooler operating modes;
- emplacement drift wall temperature below 200°C during the post-closure period to avoid potential mineralogical changes in the host rock.

The variables associated with thermally optimising the sub-surface repository design are illustrated in Figure 6 [10]. The thermal response of the repository depends on:

- the waste form thermal density profile (kW/equivalent MTHM) which is a function of the waste loading and half-lives of the immobilised radionuclides;
- the waste package thermal output (kW), which is directly proportional to the amount of waste contained in a waste package;
- the emplacement drift linear heat rate (kW/m), which is controlled by the waste package thermal output and the waste package spacing.
- the emplacement drift spacing (which combined with the amount of waste contained in each waste package and the waste package spacing dictates the area loading of the repository).

Changing these variables through design modifications and/or different operational parameters directly impacts the repository thermal regime. This methodology was used, given the thermal density of commercial spent nuclear fuel and the thermal constraints listed above, to guide the sub-surface design of the proposed repository at Yucca Mountain. This process resulted in the current sub-surface repository design including the following design parameters [9]:

- drift spacing of 81 meters;
- waste package spacing of 0.1 meters;
- average emplacement drift line load of 1.45 kW/m;
- maximum waste package thermal output of 11.8 kW;
- ventilation flow rate of 15 m^3/s;
- duration of waste emplacement of 23 years followed by 50 years of forced ventilation following emplacement of the last waste package.

The first variable shown in Figure 6, thermal output of assemblies, is equivalent to the thermal output of a waste form generated from an advanced fuel cycle. The waste form thermal output would depend on the specific radionuclides included in the waste and the waste loading (percentage of waste mass in the waste form matrix). The mass of radionuclides in the waste streams would depend on the separations process, namely those radionuclides that would be targeted for separation (*e.g.* heat producing radionuclides) and how efficient the process is at separating those radionuclides. A low separation efficiency would result in a larger amount of heat-producing radionuclides being lost to the waste and combined with high waste loadings would result in a high waste form thermal output. Contrarily, a high separation efficiency combined with a low waste form loading would lead to a lower waste form thermal output. This is shown schematically in Figure 7.

While Figure 7 shows separation efficiency and waste loading as being independent, the waste form loading would be coupled to separation efficiency. For example, a lower separation efficiency would result in a larger amount of heat-producing radionuclides being lost to the waste and thermal limits on the waste form itself could limit the waste loading.

Figure 6: Variables affecting the thermal performance of the repository [10, Figure 2-8 therein]

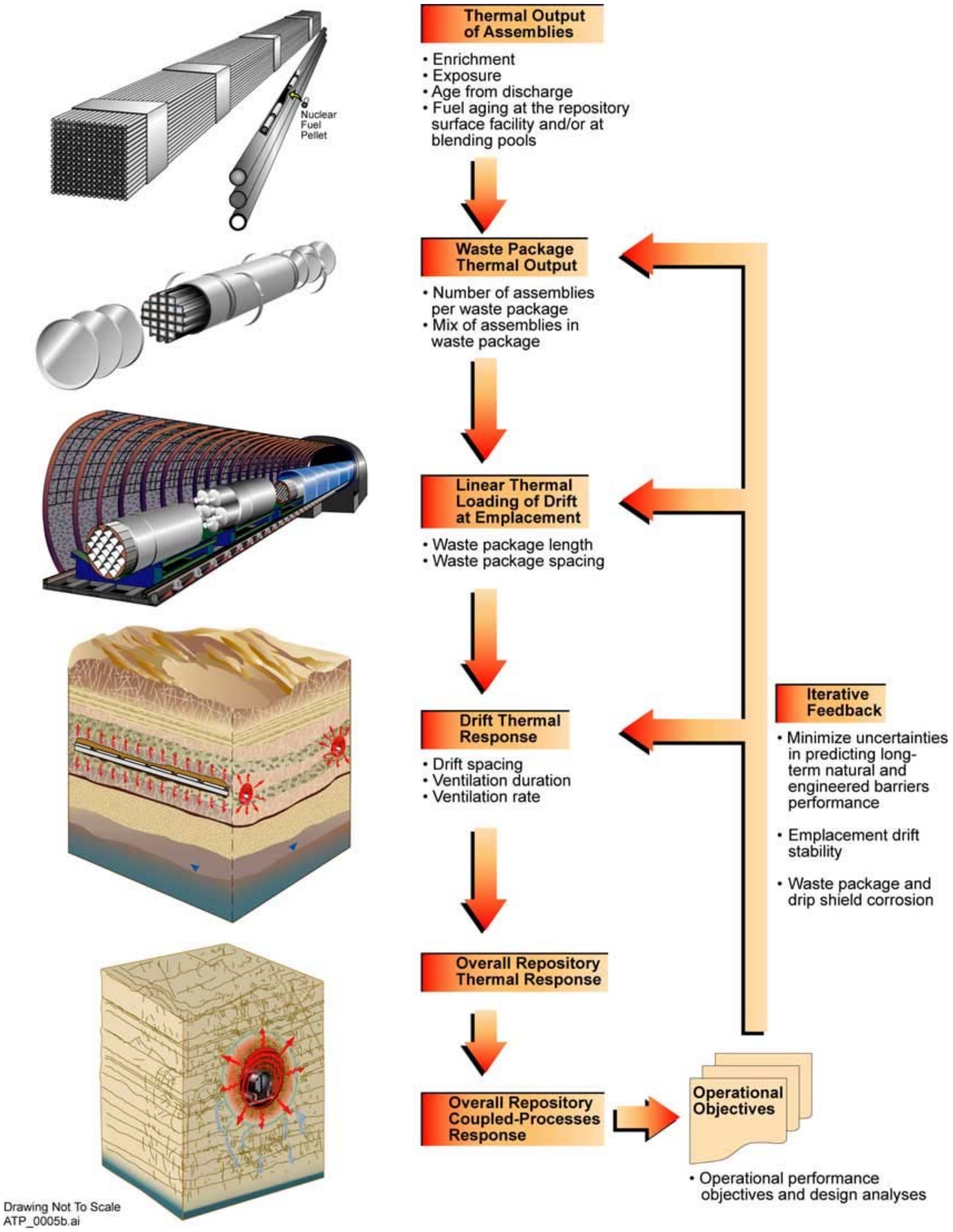
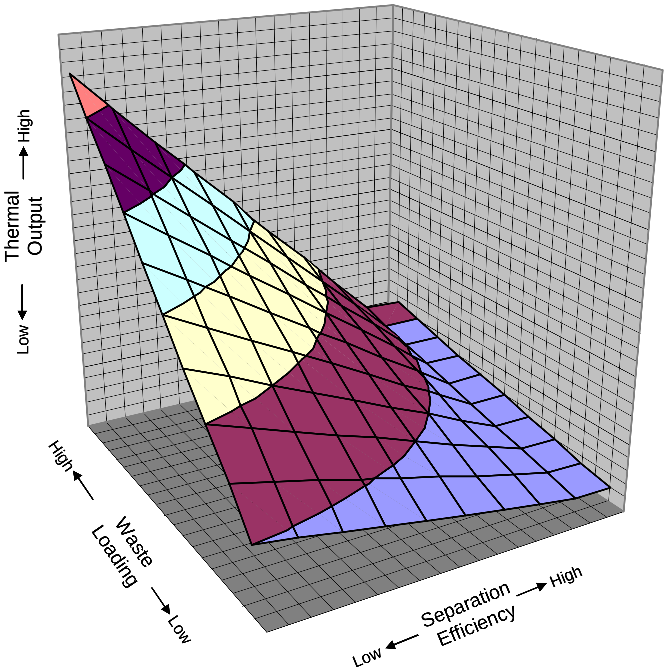


Figure 7: Schematic showing dependence of waste form thermal output, separation efficiency and waste form loading efficiency



Reducing the waste form thermal output allows increased flexibility in the optimisation of the design and operation of a repository by reducing one of the key variables affecting the repository thermal behavior. The linear thermal loading and thermal response would then depend on other design factors such as whether there is additional decay storage, how the waste is packaged and emplaced in a geologic disposal facility, and whether the facility is ventilated.

Because the recycling technology still must be demonstrated to be commercially deployed, the waste form characteristics have yet to be determined, and a specific disposal site has not been selected for the disposal of wastes from an advanced nuclear fuel cycle, it is not currently possible to conduct such optimisation analyses. Thus, the amount of waste that could be emplaced in a given area, under an advanced nuclear fuel cycle cannot presently be determined at present in any geologic media.

However, analyses have been conducted to determine the thermal benefit that could be realised with recycling and disposing of wastes in a repository in unsaturated tuff while meeting the thermal goals presented above [3]. The results demonstrated that waste could be emplaced in a geologic repository at densities at least ten times greater than that for the direct disposal of spent nuclear fuel if separation efficiencies exceeding 90% are realised.

Similar thermal benefits would result in other geologic media. Thermal criteria in disposal systems located in other geologic media are similar to those for the repository at Yucca Mountain, Nevada. For example:

- The Gesellschaft für Anlagen und Reaktorsicherheit (GRC) assumes that a 200°C limit at the container/salt interface would be established by regulation for a repository located in a salt formation in Germany [2].
- The Environmental Assessment for the disposal of spent nuclear fuel and high-level nuclear waste at the Deaf Smith County, Texas site indicates that maximum allowable repository temperature is less than 250°C [11].
- The Agence nationale pour la gestion des déchets radioactifs (ANDRA) in France has adopted a 90°C limit for the hottest point in swelling clay buffers in a granite disposal system and at any point in a disposal system in argillaceous media [12,13].

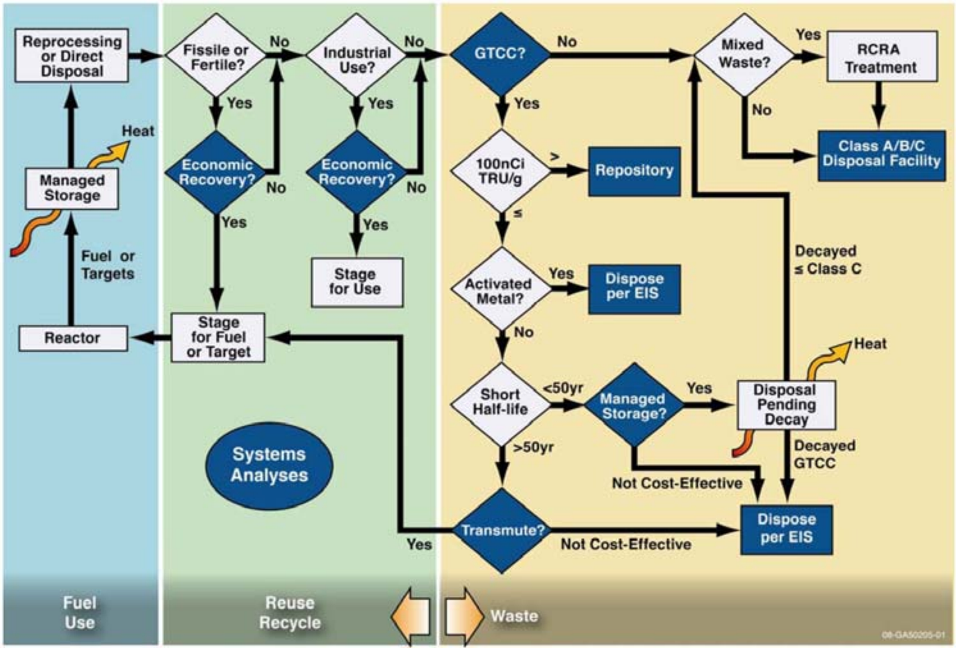
Analyses completed by the Empresa Nacional de Residuos Radioactivos SA (ENRESA, Spain) for a repository in granite show essentially no benefit in terms of improved loading density for disposing HLW-containing minor actinides (MA) as compared to the direct disposal of SNF while a factor of approximately four benefit could potentially be achieved for the disposal of non-MA bearing HLW [2]. Analyses completed by the Belgian Nuclear Research Centre SCK•CEN for a repository in clay showed similar results, but further demonstrated that the removal of the short-lived fission products in addition to the MA could potentially result in a benefit of approximately ten [2]. Since the thermal criteria for a salt media would likely be of similar magnitude to those of granite and clay and also would apply in the vicinity of the emplaced waste (as opposed to some distance into the host rock), a similar thermal response is expected.

Waste management system for an advanced nuclear fuel cycle

The benefits of minor actinide and fission product partitioning and transmutation discussed above consider only the end-state advanced nuclear fuel cycle and focused only on geologic disposal. There would likely be a transition from once-through or limited plutonium recycling fuel cycles utilising light water reactors that are in use world wide today to a closed advanced fuel cycle where plutonium and other minor actinides are transmuted in fast reactors.

In addition, the waste management system is broader than disposal in a geologic repository. This system includes the recycling facilities, the waste processing facilities, storage facilities and the waste transportation infrastructure. While protecting workers and the public (risk/safety) is of highest priority, other factors such as regulatory constraints, economics, social/institutional aspects, and environmental protection must be considered when making decisions regarding waste management under an advanced nuclear fuel cycle.

Figure 8: Global Nuclear Energy Partnership/Advance Fuel Cycle Initiative Integrated Waste Management Strategy logic diagram



Class A, B, C and Greater than Class C (GTCC) are low-level radioactive waste classification levels per US Code of Federal Regulations Title 10, Part 61. Class A, B and C low-level radioactive wastes are analogous to low-level wastes considered internationally while GTCC low-level radioactive waste is analogous to intermediate level wastes considered internationally.

EIS denotes the GTCC Environmental Impact Study currently under development by the US Department of Energy Office of Environmental Management

Mixed waste is hazardous waste defined under the Resource Conservation and Recovery Act (RCRA) that is also radioactive.

The AFCI/GNEP Integrated Waste Management Strategy

Recognising the variety of considerations involved in developing a waste management system for an advanced nuclear fuel cycle, the GNEP/AFCI programme developed an Integrated Waste Management System (IWMS) [6]. The GNEP/AFCI IWMS establishes a logical framework for directing waste management and disposition alternatives based on waste radionuclide content. While the GNEP/AFCI IWMS was established considering US laws and regulations, the overall logic is applicable to any waste management programme. The GNEP/IWMS logic is shown in Figure 8.

The GNEP/AFCI IWMS emphasises the recycle and reuse of material either as nuclear fuel or for industrial application. The decision to recycle or reuse would be based on an economic recovery evaluation factoring in the value of the material and the cost avoidance of disposal. Under the Nuclear Waste Policy Act HLW in the US is classified as highly radioactive material resulting from the reprocessing of spent nuclear fuel, and disposal in a deep geologic repository is required [14]. Rather than requiring all waste generated under an advanced nuclear fuel cycle be disposed as HLW in a geologic repository, the GNEP/AFCI IWMS considers disposal options based on the risk of the waste streams and waste forms. A key aspect of the GNEP/AFCI IWMS is the inclusion of managed storage facilities where isotopic concentrations, and heat, are allowed to decay prior to storage.

Implementation of the GNEP/AFCI IWMS through system analysis

System analysis is an important part of implementing the GNEP/AFCI IWMS. While system analysis is pointed out in Figure 8 when considering whether it is economical to reuse/recycle material, system analysis is also important in determining the optimal waste management strategy (*i.e.* processing steps, waste forms). System analyses are currently under way within the GNEP/AFCI programme to further optimise among waste disposition alternatives, considering both an end-state closed fuel cycle and during the transition from the once-through system now in place is the USA.

- The GNEP/AFCI reference UREX +1a process steps and waste streams shown in Figure 2 result in a number of independent waste streams and waste forms. An evaluation was conducted striving to minimise both the number of waste processing steps and the number of waste types. Rather than having multiple waste streams, the various waste streams were matched to applicable waste forms based on chemistry. The evaluation recommended the combination of metallic and oxide waste streams into two composite waste forms as shown in Figure 9. While no processing steps were eliminated, some of the waste streams were combined and the number of waste forms was reduced.

- An evaluation was just completed regarding the disposition of the transition metal fission products (TMFP) [15]. Combining the TMFP with the Cs/Sr/Ln waste stream would allow one step in the UREX +1a process to be eliminated (FPEX; Cs/Sr would be removed in the TRUEX step). The volume of the resultant glass waste form that would have to be disposed would increase. Though disposal costs would increase, the results indicate that there would be an overall cost savings. Further analysis showed that combining all of the primary process wastes (UDS, Tc, Cs/Sr, TMFP, Ln, but not cladding/hardware or volatiles) in one glass (Figure 10) resulted in the most cost-effective waste management strategy. Overall waste form volume was greater, but costs were much lower without the capital and operating costs of additional waste processes.

- The GNEP/AFCI programme is performing a system analysis study evaluating the alternatives for minor actinide management under a scenario that transitions from recycling plutonium in light water reactors to full transuranic recycle in fast reactors. The question is whether to store the minor actinides for future transmutation in fast reactors when they become available or to dispose the minor actinides in a deep geologic repository during the transition period.

- As discussed above, the removal of key heat-producing radionuclides (*i.e.* ^{241}Am, ^{137}Cs, ^{90}Sr) would reduce the thermal output of the waste that would be disposed and allow for flexibility in repository design while meeting thermal design criteria. While ^{241}Am could be transmuted in fast reactors, ^{137}Cs and ^{90}Sr would still have to be managed. The GNEP/AFCI programme is conducting a system analysis of the various alternatives for managing these heat-producing radionuclides including extended aging of spent nuclear fuel, long-term decay storage (at the recycling plant, at an interim storage facility, or at the repository), or direct disposal. Facility capital, maintenance and operating costs are being considered in addition to transportation, handling, processing, security and permitting issues.

Figure 9: Evolution of the GNEP/AFCI reference waste streams and waste forms

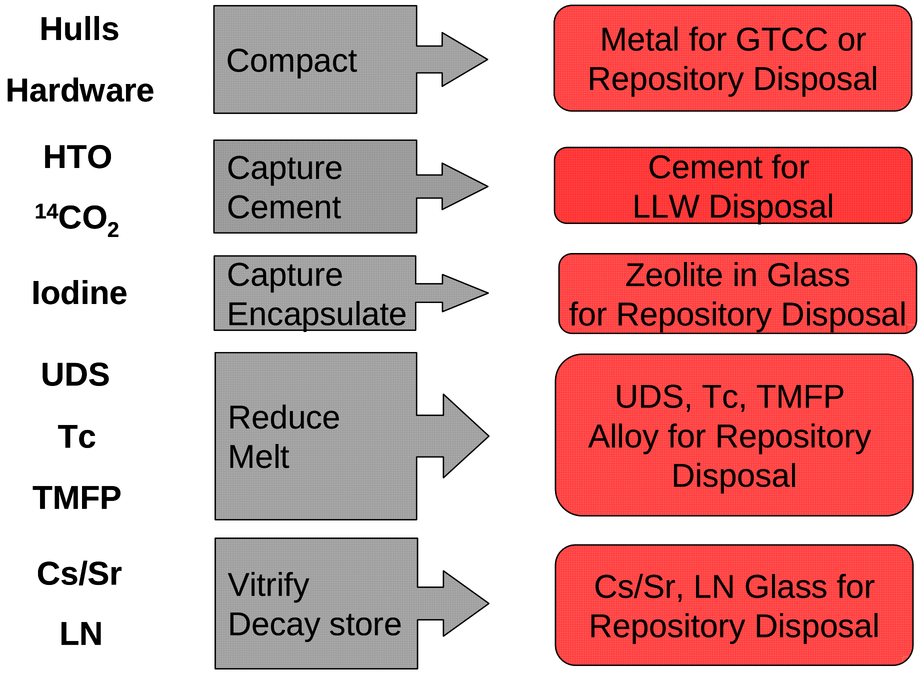
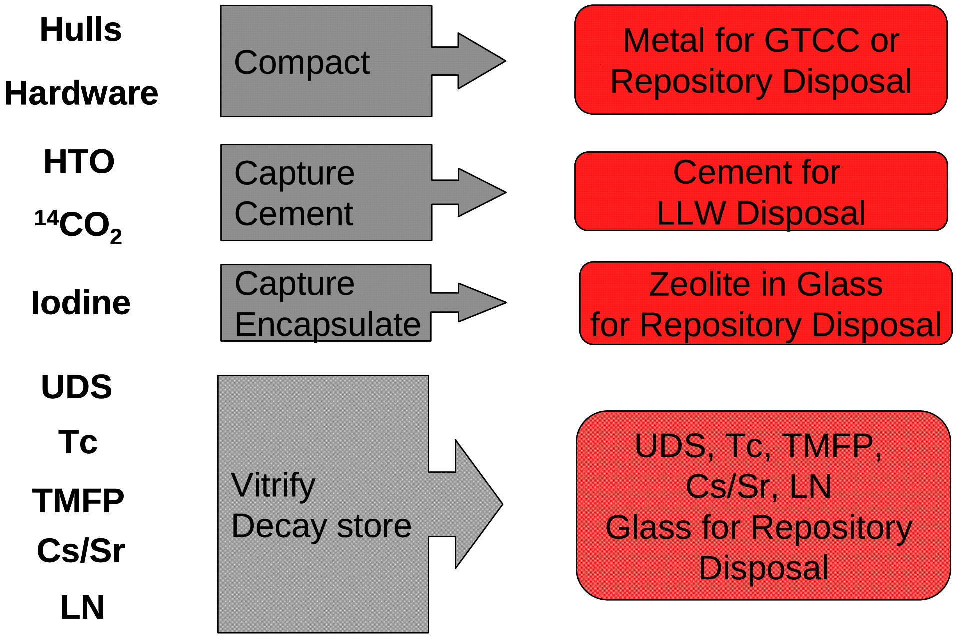


Figure 10: GNEP/AFCI waste forms resulting from further combination of waste streams



Waste form development

In addition to conducting system analyses related to waste management and disposition, the GNEP/AFCI programme is currently investigating the development and characterisation of potential waste forms that could be used to sequester the wastes that would be generated under an advanced nuclear fuel cycle. Waste forms are being selected to match waste stream chemistry and potential disposal system environments, using naturally occurring materials to guide selection of potential waste forms. Many of the waste processing steps are based on mature technologies that can be advanced and applied. No technical issues have been identified that would indicate that wastes could not be effectively isolated.

- An initial technology readiness assessment (TRA) on each of the potential waste forms for isolating wastes from an advanced nuclear fuel cycle is being conducted. The concept was developed by the US National Aeronautic and Space Administration and is being adopted by the GNEP/AFCI programme to help guide future waste form development and characterisation activities. A TRA generates a technology readiness level that indicates the maturity of a waste development process and the waste form itself. The TRA process also develops a technology maturation plan required to bring the technology to the level required for implementation.
- The GNEP/AFCI programme is developing testing protocols pertaining to the variety of waste forms that could be developed under an advanced nuclear fuel cycle. Testing will determine how the waste forms will perform in geologic repository relevant environments. Test method selection and testing protocols are based on consensus standards such as those established by the ASTM and the American Nuclear Society.
- Investigations are under way to determine processes and waste forms for sequestering volatile radionuclides released from the voloxidation and dissolution steps. In particular, the capture of iodine on silver-mordenite and other zeolitic materials is being investigated.
- Immobilisation of technetium, un-dissolved solids and the transition metal fission product waste streams in zirconium-iron alloys is being researched. It is envisioned that some of the zircaloy cladding hulls would be used to create this waste form. Both the characteristics of the waste form (loading density, phase diagram) and its long-term performance (corrosion) are being studied.
- Immobilisation of the Cs/Sr waste stream in glass and glass/ceramic materials as both a storage form and a waste form (for disposal) is being investigated. The ability to isolate the lanthanides alone, combined lanthanide – Cs/Sr, and combined lanthanide – Cs/Sr – transition metal fission products into glass waste forms is also being investigated.
- Electro-chemical processing of fast reactor spent nuclear fuel is being investigated under the GNEP/AFCI programme. Correspondingly, potential forms that could be used to isolate the wastes that would be generated are also being researched. These waste forms include a metallic alloy analogous to that discussed above and glass-bonded sodalite.

Conclusion

Advanced separations techniques being developed under the US Global Nuclear Energy Partnership/ Advanced Fuel Cycle Initiative (GNEP/AFCI) programme integrated with advanced waste form production and performance provides opportunities for flexibility and ultimately fuel cycle system optimisation. Removal of uranium and transuranics for destruction in advanced reactors while generating electricity and management of fission product waste forms in decay storage as part of a closed fuel cycle would impact the potential technical capacity of a geologic repository. Studies show that the volume of high-level waste requiring disposal in a geologic repository could potentially be reduced by 90% versus directly disposed spent nuclear fuel. Far more important than reducing volume is: *i)* recycling the actinides for energy recovery and transmutation; *ii)* decay storage to manage heat from the HLW prior to final disposition. This strategy makes the repository temperature constraints non-limiting, allowing more efficient space utilisation in the repository. An added benefit is the potential reduction of long-term risk of the waste by removing the actinides.

Acknowledgements

The authors would like to acknowledge the US Department of Energy, Office of Nuclear Energy and the Global Nuclear Energy Partnership/Advanced Fuel Cycle Initiative programmes which supported this work.

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Studies on separation of actinides and lanthanides by extraction chromatography using 2,6-bistriazinyl pyridine

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Abstract

2,6-bis(5,6-dipropyl-1,2,4-triazin-3-yl)pyridine (nPr-BTP) was impregnated on XAD-7 resin and the extraction performance of this nPr-BTP/XAD-7 resin was investigated for the uptake of Am(III) and some lanthanides from acidic nitrate solutions. The uptake behaviour of Am(III) and the lanthanides La(III), Ce(III), Nd(III), Eu(III) and Gd(III) was studied by batch experiments. It was found that the resin exhibited significantly high extraction and selectivity for Am(III) over the lanthanides. Based on the results obtained from batch studies, the separation behaviour of Am(III) from Eu(III) was examined by extraction chromatography using a column packed with the resin. A complete separation between Am(III) and Eu(III) was achieved in the column experiment.

Introduction

Nuclear energy generation gives rise to highly radiotoxic waste. One of the main concerns of the nuclear industry today is the safe disposal of high-level waste (HLW) generated during the reprocessing of the spent nuclear fuel. The toxicity of the waste is mainly due to the long-lived minor actinides such as ^{241}Am, ^{243}Am, ^{245}Cm, ^{237}Np and unrecovered Pu. The high-level waste is deposited in deep geological repositories; however, this practice is a matter of environmental concern due to the long-term radiotoxicity associated with the minor actinides. In this regard, partitioning and transmutation (P&T) is a strategy that envisages the complete removal of minor actinides from the HLW solution followed by their transmutation to other stable or low-level active nuclides by neutron bombardment in accelerators or fast-breeder reactors, thus considerably reducing the long-term radiological hazards associated with the waste.

However, lanthanides, which are also present in the HLW and are 20 times more abundant than the actinides, preferably absorb the neutrons thereby affecting the transmutation efficiency of the actinides and thus it becomes imperative to separate them from the lanthanides. This separation is difficult due to the similar chemical properties of both. However, it has been observed that there is a modest enhancement of covalency in the actinide-ligand bonding compared to lanthanides and hence ligands containing soft-donor atoms such as nitrogen and sulfur form stronger bonds with actinides relative to the lanthanides [1]. Based on this approach, several new extractants have been developed to particularly extract the trivalent actinides and achieve the lanthanide-actinide separation [2-5].

One promising class of extractants is the 2,6-bis(5,6-dialkyl-1,2,4-triazin-3-yl)pyridines (BTP), which were first reported by Kolarik, *et al.* [6,7]. Besides giving good separation factors, the BTP also have the advantage of being CHON extractants and therefore are completely incinerable. With regard to a liquid-liquid extraction process, the problem associated with the use of BTP was the need to use a phase modifier such as 1-octanol to enhance its solubility in the hydrocarbon diluents. Therefore, instead of using liquid-liquid extraction, an attempt has been made in the present work to use these extractants through extraction chromatography.

The resin for extraction chromatography was prepared by impregnating 2,6-bis(5,6-dipropyl-1,2,4-triazin-3-yl)pyridine (nPr-BTP) on XAD-7 resin and has been evaluated with respect to the uptake of Am(III) and some lanthanides. Batch studies were carried out to investigate the uptake behaviour of Am(III) and the lanthanides La(III), Ce(III), Nd(III), Eu(III) and Gd(III). Column studies were carried out to determine the separation behaviour of Am(III) from Eu(III) using the extraction resin. The results of the above experiments are reported in this paper.

Experimental

Preparation of nPr-BTP

2,6-bis(5,6-dipropyl-1,2,4-triazin-3-yl)pyridine (nPr-BTP) was synthesised through the reaction between 2,6-pyridinedicarboxamide dihydrazone and octane-4,5-dione according to the synthesis procedure reported by Case [8]. Octane-4,5-dione was synthesised by oxidising trans-4-octene with potassium permanganate according to the procedure reported by Sharpless, *et al.* [9]. The BTP so formed has been characterised using IR, MS and CHNS techniques. The absence of carbonyl peak in IR and molecular ion peak of 406 corresponding to MH+ in mass spectra indicates the synthesis of nPr-BTP.

Preparation of extraction resin

For impregnating 2,6-bis(5,6-dipropyl-1, 2,4-triazin-3-yl)pyridine on the XAD-7 resin, the XAD-7 resin (supplied by Thermax Ltd., Pune) was washed well with methanol and then with acetone and finally dried in air. An amount of 16 g of the nPr-BTP ligand was placed in a round-bottom flask and dissolved in a minimum amount of dichloromethane as diluent. The dried XAD-7 resin (24 g) was added to the solution and the slurry was rotated vigorously for 2 h at 298 K. The diluent was removed at 323 K using a rotary evaporator. The resin was dried in air. The weight increase in resin indicates the loading of BTP by 40%.

Batch studies

The uptake of Am(III) and the lanthanides La(III), Ce(III), Nd(III), Eu(III) and Gd(III) was studied by equilibrating a known amount of air-dried resin (0.25 g) with a known volume (3 mL) of the HNO_3-NH_4NO_3 solutions of desired concentration in a temperature-controlled water bath at 303 K for three hours. The effect of the HNO_3 concentration on the uptake of Am(III) both in the presence and absence of NH_4NO_3 was also studied. The effect of the HNO_3 concentration on the uptake of the above-mentioned lanthanides in the presence of NH_4NO_3 was also investigated. Further, the effect of NH_4NO_3 concentrations on the uptake of Am(III) and the lanthanides was studied. The kinetics of Am(III) uptake by the extraction resin was also investigated.

An assay of ^{241}Am was carried out by gamma counting in a NaI scintillation counter. The lanthanide concentrations were estimated using ICP-MS. The distribution coefficient was calculated by the following equation:

$$K_d = \frac{C_0 - C}{C} \times \frac{V}{W}$$

where C_0 and C denote the element or nuclide concentration (activity) in the aqueous phase before and after equilibration respectively, V is the volume of the aqueous phase used (mL), and W indicates the weight of the extraction resin (g).

Column runs for Am(III) and Eu(III) separation studies

A glass chromatographic column with a 10-mm inner diameter was packed with 2 g of resin. Column operations were carried out at room temperature. The bed volume as calculated from the column dimensions was found to be 6 mL. Prior to the introduction of the sample solutions, the column was preconditioned by passing three column volumes of 2 M NH_4NO_3-0.1 M HNO_3. A solution of 2 M NH_4NO_3-0.1 M HNO_3 (50 mL) spiked with Am(III) and Eu(III) (1:40 ppm) was passed through the column. The flow rate was maintained at 0.5 mL/min. Am(III) was eluted with 0.3 M DTPA solution adjusted to a pH of ~4. The loading and elution profiles for Eu(III) and Am(III) were obtained by plotting radioactivity per unit volume of different volume increments of the mobile phase vs. volume of mobile phase passed.

Results and discussion

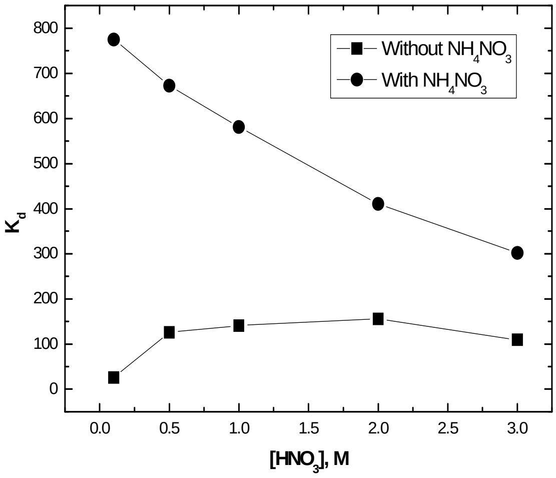
Lanthanide-actinide extraction studies by batch uptake

Figure 1 compares the distribution coefficient (K_d) values for the extraction of Am(III) by nPr-BTP/XAD-7 resin with and without NH_4NO_3 from nitric acid solution of different concentrations. It is seen from the figure that K_d values for the extraction of Am(III) from nitric acid, without ammonium nitrate are less than 200, whereas with ammonium nitrate the K_d values are significantly higher. The uptake of americium from nitrate solution onto nPr-BTP impregnated extraction resins is presumed to take place in accordance with the following equation:

$$M^{3+} + 3NO_3^- + n(\mathrm{nPr-BTP}) = M(NO_3)_3(\mathrm{nPr-BTP})_n \qquad (1)$$

The increase of Am(III) adsorption with increasing nitrate concentration can be explained by the adsorption equilibrium represented by Eq. (1). The presence of NH_4NO_3 greatly enhances the extraction of Am(III) by the resin and this result agrees well with that reported in liquid-liquid extraction by Kolarik, *et al.* [6,7] for the extraction of Am(III) and Eu(III). Maximum uptake of Am(III) has been observed from 0.1 M HNO_3 containing 2 M NH_4NO_3. The decrease of the distribution coefficient values with increasing acid concentration can be ascribed mainly to the increasing protonation of the extractant. It is reported that BTP is a tridentate extractant, forming 1:3 complexes with the trivalent metal ion, where the three BTP ligands are bound to the M(III) ion through three nitrogen atoms. Hence, the metal centre is nine co-ordinate, each BTP ligand being attached via the nitrogen atom of the pyridine fragment and the nitrogen atoms at the two-position of the triazine rings [10]. Protonation of the extractant will render these nitrogens unavailable for co-ordination thereby decreasing the extraction ability at higher acid concentrations. Further nPr-BTP extractant is also liable to dissolve in

Figure 1: Effect of nitric acid concentration on the uptake of Am(III) by nPr-BTP/XAD-7 resin with and without 2 M NH_4NO_3 (0.25 g nPr-BTP/XAD-7, 303 K, 3 h)



concentrated acid because of the protonation and it is reported that the leaching of the nPr-BTP extractant from the resin increases with increasing acid concentration [11-13]. The uptake of the lanthanides La(III), Ce(III), Nd(III), Eu(III) and Gd(III) by nPr-BTP/XAD-7 resin at different acidities in presence of 2 M NH_4NO_3 was studied (Figure 2) and it is clearly seen that the lanthanides are not extracted by the resin at any acidity. It is seen from Figure 1 that the K_d value for the extraction of Am(III) is maximum at 0.1 M nitric acid in the presence of ammonium nitrate. Hence the effect of ammonium nitrate concentration was studied at 0.1 M nitric acid. The results are presented in Figure 3 [14]. It is seen that the distribution coefficient (K_d) value of Am(III) increases with increase in NH_4NO_3 concentration. This increase can be explained by Eq. (1). However, for Eu(III) the increase in K_d values is observed to be very small, whereas for other lanthanides there is no increase in K_d values at all NH_4NO_3 concentrations. Thus, nPr-BTP/XAD-7 displays high selectivity for Am(III) compared to trivalent lanthanides. Table 1 [14] shows the separation factor values for the separation of americium from the lanthanides. The values for Eu were found to be much less than those for the others. However, these values are adequate for a successful separation of the two elements in a chromatographic column. For our studies we kept the NH_4NO_3 concentration constant at 2 M in 0.1 M HNO_3, and have been able to demonstrate a good separation performance of the extraction resin in the column studies.

Figure 2: Effect of nitric acid concentration on the uptake of Am(III) and lanthanides by nPr-BTP/XAD-7 resin (0.25 g nPr-BTP/XAD-7, 2 M NH_4NO_3, 303 K, 3 h)

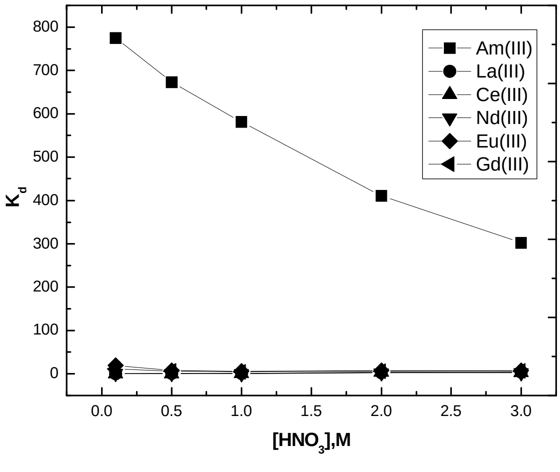


Figure 3: Effect of nitrate concentration on the uptake of Am(III) and lanthanides by nPr-BTP/XAD-7 resin (0.25 g nPr-BTP/XAD-7, 0.1 M HNO_3, 303 K, 3 h)

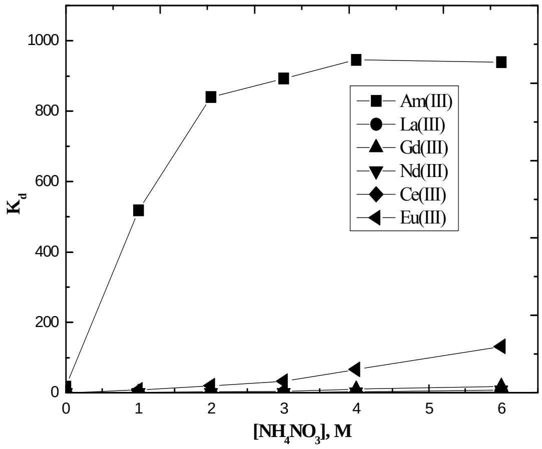
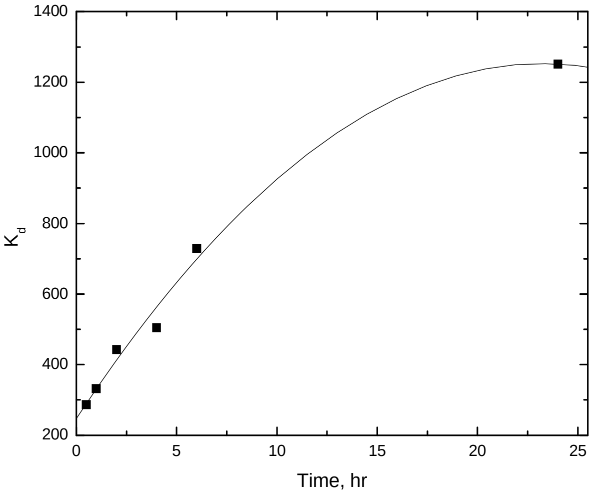


Table 1: Separation factors for americium-lanthanide separations (0.25 g nPr-BTP/XAD-7, 0.1 M HNO_3, 303 K, 3 h)

$[NH_4NO_3]$, M	Separation factor				
	La	Ce	Nd	Eu	Gd
0	192	173	173	173	173
1	2 730	5 187	5 187	66	451
2	2 471	7 638	1 400	43	296
3	8 925	8 926	8 926	27	246
4	1 412	937	347	14	92
6	770	465	117	7	52

The rate of absorption of Am(III) from 0.1 M nitric acid and 2 M ammonium nitrate solution was studied by measuring K_d values for its extraction at different intervals of time. It is seen from Figure 4 the distribution coefficient values increased with the increasing time of equilibration and equilibrium is reached in 24 hours. The equilibrium with other metal ions was not studied, as it is assumed to be similar for other metal ions also. For our K_d measurements we have equilibrated for three hours.

Figure 4: Kinetics of the uptake of Am(III) by nPr-BTP/XAD-7 resin (0.25 g nPr-BTP/XAD-7, 0.1 M HNO_3, 2 M NH_4NO_3, 303 K)



Column studies for the separation of Eu(III) and Am(III)

For column run, a solution of 2 M NH_4NO_3-0.1M HNO_3 (50 mL) spiked with Am(III) and Eu(III) (1:40 ppm) was passed through the column. As expected from the results of the batch experiments, it was observed that europium was not retained in the column and up to 99.4% of the Eu was recovered at the loading stage itself. Up to 90% of the Am was retained in the column. The loaded Am(III) was then recovered by passing 0.3 M DTPA solution (pH = 4.0) and 60% of Am was recovered within the first three column volumes and further tailing was observed. The presence of Eu in the eluted Am from the BTP-XAD7 column was analysed using High Resolution Gamma Spectrometry, and the amount of Eu present in the Am was found to be only 0.6%, thereby clearly indicating a clear separation of the europium from americium. Further efforts are on to reduce the tailing.

Figure 5: Loading of Am(III) and Eu(III) onto a column of nPr-BTP/XAD-7

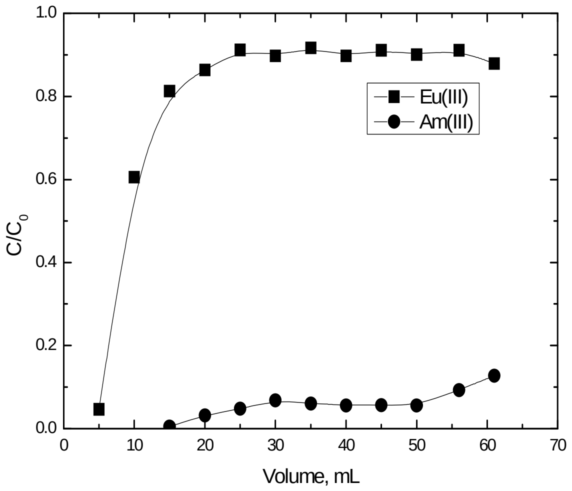
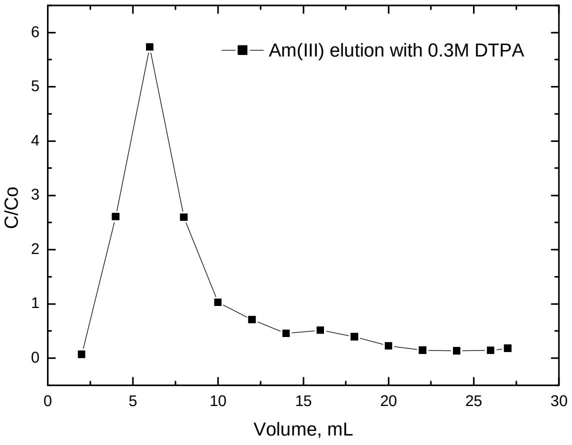


Figure 6: Elution of Am(III) from nPr-BTP / XAD-7 column with 0.3 M DTPA



Conclusion

2,6-bis(5,6-dipropyl-1,2,4-triazin-3-yl)pyridine (nPr-BTP) was impregnated onto XAD-7 resin. The resin displayed significantly high selectivity for americium and good separation factors for the separation of other lanthanides from the same. Column runs for the separation of americium from europium gave good results with 99.4% Eu being removed in the loading stage itself. The elution of Am from the column using DTPA was found to be 60% and efforts are on to improve the same.

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Recent development of pyrochemical processing and metal fuel cycle technology in CRIEPI

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Abstract

This paper summarises the activity of pyroprocessing and metal fuel development during the past several years in CRIEPI. Process developments of electrorefining, reductive extraction to recover actinides in salt and electrochemical reduction are described with technological developments by use of engineering scale model. Regarding P&T activity, a demonstration test of TRU recovery from genuine HLLW and post-irradiation examination of U-Pu-Zr-MA-RE fuel discharged from the Phénix reactor are underway. The results of P&T technology and metal fuel cycle technology will be reviewed in the framework of OMEGA programme and FaCT programme, respectively.

Introduction

The recent steep rise in uranium ore price clearly show the future demand on the nuclear power as CO_2 emission free energy supply. However, breeding of plutonium by fast reactor is necessary to sustain long-term nuclear energy supply because uranium ore has a certain limit. On the other hand, use of plutonium usually faces the matters of nuclear proliferation, high-fuel cycle cost, build-up of radioactive waste of long half-lives, etc. The metal fuel fast reactor cycle is one of the most attractive technologies to answer these matters. The nature of metal fuel enable us to employ pyrometallurgical reprocessing which has an intrinsic proliferation-resistant feature due to the inherent difficulty of extracting weapons-usable Pu. Long-lived transuranium elements are recovered together with Pu in an electrorefining step, and are served for fuel fabrication to be transmuted in the fast reactor. Combination of the pyrometallurgical reprocessing technology and the injection fuel casting technology offers substantial reduction of fuel cycle cost compared with the conventional wet reprocessing and the pellet fabrication system. Hence CRIEPI has been studying the metal fuel fast reactor cycle since 1987 as an outstanding alternative in the future energy source. Systematic study has been carried out in the framework of the IFR programme before 1994 [1], and CRIEPI has been continuously involved to develop pyroreprocessing technology since 1985 by participating in the IFR programme and after the end of the programme as well. CRIEPI's activity has been focusing not only on an aspect of metal fuel cycle but also on P&T of long-lived radioactive nuclides [2], in which includes a process to convert nitrate solution to chloride form [3]. The pyroprocessing of oxide fuels to introduce pyroreprocessing with electrorefining has been investigating by use of electrochemical reduction technology [4].

This paper describes an overview of the progress of our metal fuel cycle project, including our partitioning and transmutation (P&T) programme. The results regarding the P&T programme will be reviewed in the framework of the Japanese OMEGA programme. The most important result is the pyropartitioning test carried out as a joint study with JRC-ITU to recover TRU from real HLLW. Collaborations with other organisations play an effective role to establish the process and to develop an engineering scale of installation in this programme.

Concept of closing actinide fuel cycle with metal electrorefining

The metal electrorefining with dual cathodes, one for uranium deposition and the other for actinide collection into liquid cadmium, is an essential device to separate uranium as well as transuranium elements [5]. Figure 1 represents the process flow of pyroreprocessing. The electrorefining and cathode processing for salt/cadmium distillation are the key devices to reproduce a fissile material for fabricating metal fuel. Reductive extraction is another essential process to harvest actinides from salt, in which fission products and actinides are accumulated during electrorefining operation, which contributes to waste volume reduction and minimisation of TRU losses to waste for disposal. The electrochemical reduction has a high potential to introduce conventional oxide fuels into the metal fuel cycle. The pyroreprocessing described in this paper has outstanding advantages. It can treat short-cooled fuels or high TRU content fuels like MOX without degradation of liquid media due to radiation and acid, because molten salt and liquid metal are used instead of organic material and nitric acid. Milder control on criticality restriction should be allowed because of the lack of an aqueous phase. The closer electrochemical potentials of actinide, U, Np, Am, probably Cm [6] in chloride salt/liquid cadmium does not allow an easy separation of plutonium with high enough purity to make weapons-grade material.

Development of pyroreprocessing for metal fuels and oxide fuels

In order to develop a process flow sheet reliably enough to evaluate industrial applicability, several experiments to measure detailed material balances have been carried out. Pyrochemical process in sequential mode was tested by CRIEPI-JAEA joint study shown in Figure 2 [7]. MOX fuels were reduced to metal form by electroreduction, and the products were charged in the electrorefiner to recover U and Pu-U as cathode deposits. The deposits were heated up for distillation of cadmium and salt to obtain U or U-Pu alloy. The obtained mass balance is enough to demonstrate the feasibility of the process in sequential mode. Tests to recover actinides from irradiated metal alloy fuels are planned in the next phase as joint study with JRC-ITU shown in Figure 3.

Figure 1: CRIEPI's pyrochemical process for recycle of actinides

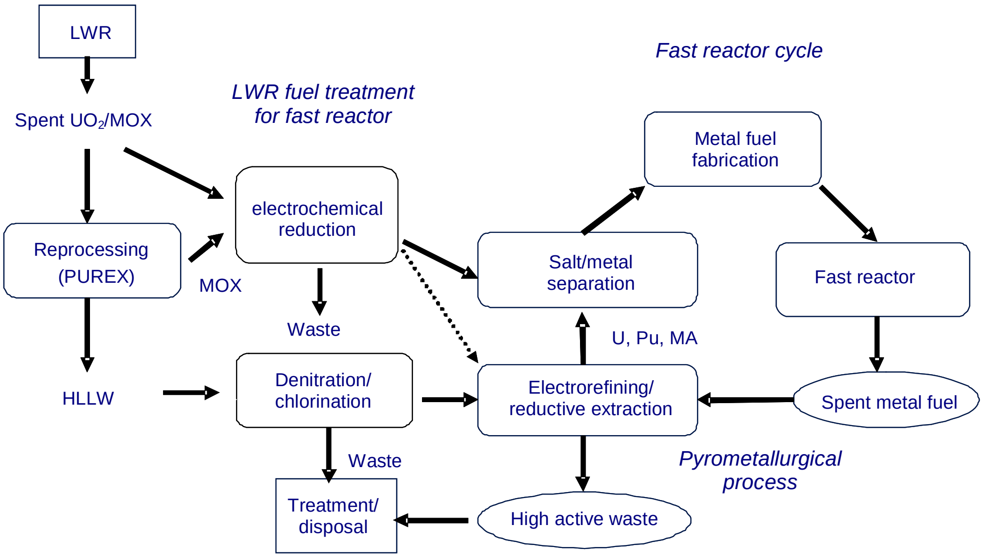


Figure 2: Pu glove box for a series verification of pyrochemical reprocessing

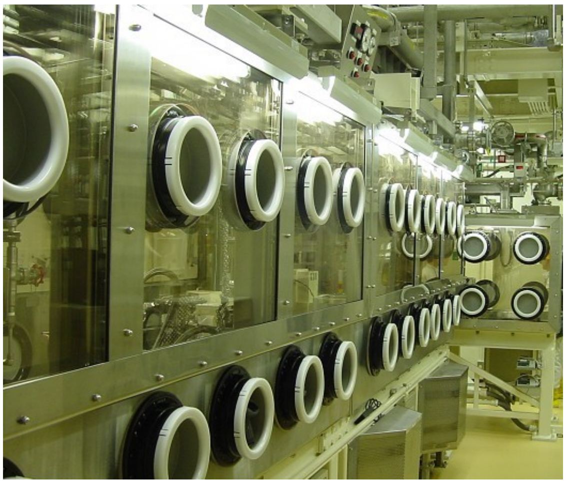


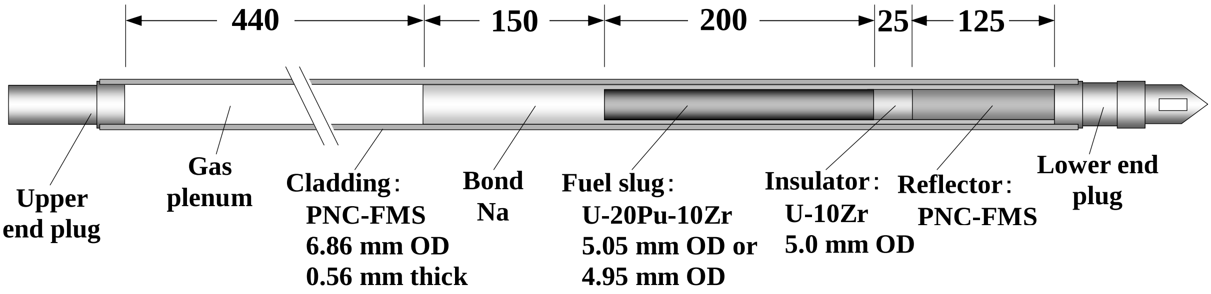
Figure 3: Ar hot cell for pyroprocess development

Development of the engineering technology is a key issue for industrialisation. Electrodes have been developed for improving throughput of electrorefiner. The anode and solid cathode pair, called high-throughput electrorefiner module, was developed. Deposited uranium crystal on the cathode is periodically scraped off for recovery [8]. After the many improvements, throughput of 789 g-U/h was achieved in a module. As for the liquid Cd cathode module, high-temperature liquid transport technology was developed to move cathode Cd product to the cathode processor. Engineering-scale electrorefiner of 780 mm internal diameter was installed in large Ar glove box, and feasibility of the Cd cathode with liquid transportation was demonstrated with using actinide-simulating elements [9]. A three-stage molten salt/liquid metal contactor for Ln/An separation was developed and the separation was demonstrated using Ce, Gd and Y as substitutes of U and Pu or MA, and other lanthanides [10].

Development of metal fuel

Though more than a thousand pins of U-Zr or U-Fissium were irradiated in EBR-II and FFTF, only several hundreds pins with U-Pu-Zr had been irradiated mostly in EBR-II. In order to verify their viability for commercial application, irradiation experiences with wider irradiation condition are necessary. The metal fuel irradiation programme has started as joint study with JAEA. U-Pu metal has been prepared by electrochemical reduction of MOX pellets. Fuel composition was adjusted to U-20wt.%Pu-10wt.%Zr by casting uranium and zirconium metal with U-Pu metal. Based on experience with engineering scale U-Zr injection casting furnace [11], U-Pu-Zr fuel single pins were injection casted into an Ar glove box, and fabricated as fuel element shown in Figure 4 [12]. The composition, homogeneity and impurities of the alloy are under analyses with appropriate means. Test fuel pins are under fabrication for next irradiation campaign at JOYO fast reactor.

Figure 4: U-Pu-Zr fuel pin fabricated for JOYO irradiation

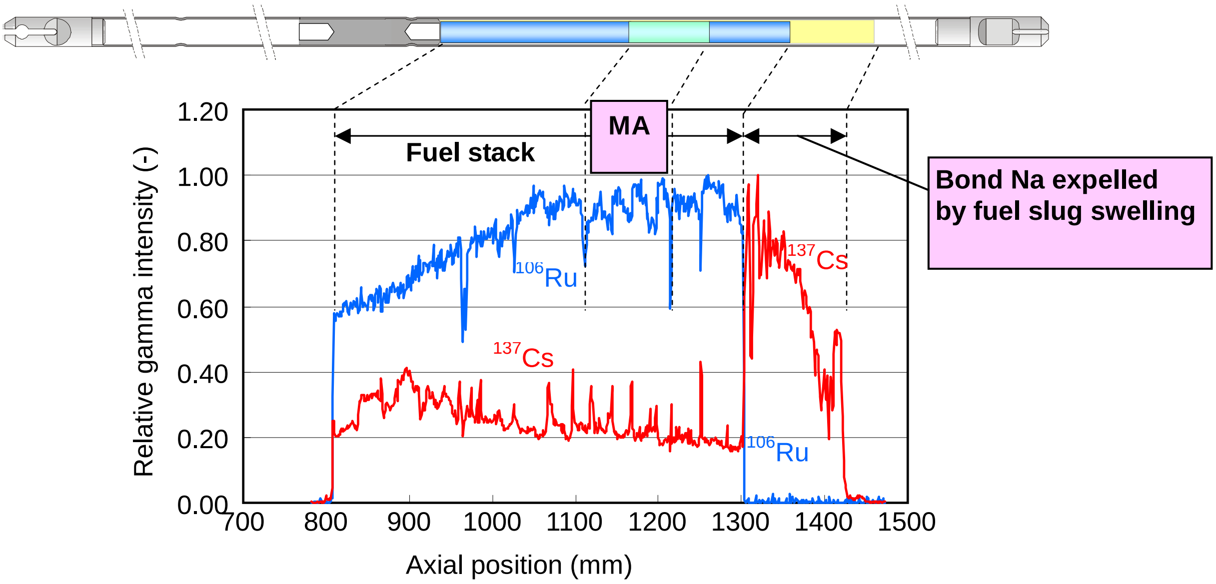


Partitioning and transmutation study

The MA concentrated HLLW prepared from PUREX raffinate was dried at around 100°C, and calcined at around 500°C under air flow. According to the ICP-MS analysis, only 0.3% of Ru compared with the initial amount was trapped in the scrubber. So far almost all of the fission products and the actinides are expected to have remained in the denitrated material. The material was then charged in a graphite crucible together with LiCl-KCl eutectic salt in an Ar atmosphere hot cell. The crucible was heated at around 650°C under chlorine gas atmosphere. The chlorinated product was sampled to dissolve in water for ICP-MS analysis. Nearly 100% of actinides in HLLW were found to be recovered as chlorinated product. It will be served for demonstration test of liquid-liquid extraction [13]. Separation factors and distribution factors will be measured for the extraction. These experimental data will be compared with equilibrium distribution data obtained with unirradiated actinides [14].

The Phénix irradiation programme for U-Pu-Zr with minor actinides, the so-called METAPHIX programme is being carried out as joint study with JRC-ITU. Three different compositions of metal fuel pins were prepared for this programme. U-19Pu-10Zr with 2% MA and 2% RE, with 5% MA and 5% RE, and without MA and RE were fabricated, where MA means Np, Am and Cm, and RE denotes Ce, Nd, Y and Gd. Compositions of MA and RE are decided based on the spent LWR fuel composition calculated by the ORIGEN-II code. The MA metals were obtained through chemical reduction of each oxide with metal reductant such as Nd or Th followed by vaporisation/deposition treatment. Three pins with different compositions are loaded in the same irradiation capsule. Three capsules were charged in Phénix to achieve three different burn-ups. Irradiation has started since early 2004 at the Phénix fast reactor under the collaboration with CEA. Three capsules with three fuel pins were discharged, achieving 2.5 at.%, 7.0 at.% and 11.0 at.% of burn-up, respectively. Non-destructive analyses were carried out for lower burn-up fuel pins as shown in Figure 5 [15]. The behaviour agreed well with the irradiation behaviour of usual U-Pu-Zr fuels. The first three pins were transported to JRC-ITU and under destructive analyses to measure fission gas release, metallography, MA transmutation rate and element analysis along radial and axial direction. Then recovery of actinides by molten salt electrorefining will be tested with the irradiated METAPHIX fuels. Subsequently, pins with intermediate and high burn-ups will be transported to serve for the same study. Prior to hot operations, experiments with unirradiated fuels were carried out. Behaviour of anodic dissolution of U-Pu-Zr fuels in LiCl-KCl molten salt are clarified for the first time [16]. Recovery and separation of MA on liquid Cd cathode is also clarified using U-Pu-Np-Am-Cm-RE-Zr fuel [17]. The same method will be applied for the irradiated METAPHIX fuels.

Figure 5: U-Pu-Zr test piece fabricated for JOYO irradiation



Conclusion

This paper summarises the activity of pyroprocessing and metal fuel development over the past several years in CRIEPI. Elemental technology of electrorefining, reductive extraction to recover actinides in salt, electrochemical reduction and fuel fabrication have been verified to apply on the reprocessing of spent metal and oxide fuels from points of process development and engineering scale application. Based on these achievements, flow sheet and design study will be re-evaluated for the fast breeder reactor cycle technology development (FaCT) carried out by JAEA, electric power companies and CRIEPI. The P&T activity will be reviewed in the framework of the OMEGA programme.

Acknowledgements

We would like to acknowledge these activities have been successfully achieved through strong collaboration with JAEA and JRC-ITU. Parts of this work are the results of the following programmes: “Development and improvement of electrorefining process”, “Development of engineering technology basis for electrometallurgical pyroprocess equipment”, “Application of electrochemical reduction to pyrochemical reprocessing for oxide nuclear fuel”, entrusted to CRIEPI by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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Partitioning of fission products and a waste salt minimisation during a pyroprocess*

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Abstract

KAERI is developing technologies which could reduce the increasing amount of spent fuel and dramatically decrease the disposal load, through recycling a waste salt in a pyroprocessing system. This study aims at providing a new way to minimise the waste salt to be disposed of, while removing the fission products generated during a pyroprocessing procedure. The main pyroprocessing processes being developed by KAERI include a voloxidation for pulverising an oxide fuel pellet to an oxide powder, an electroreduction for converting an oxide powder into a metal by using LiCl salt, an electrorefinining for recovering uranium from the converted metal by using a LiCl-KCl eutectic mixture and a waste treatment for treating the fission products arising from these processes. KAERI is presently performing a study on the removal of Cs, Sr and lanthanides including Y from a pyroprocess. First, KAERI has been testing various technologies for a partitioning of Cs and Sr from a spent nuclear oxide fuel. Among them, a typical method is to remove Cs in the form of an oxide gas from the voloxidation process and then to remove the remaining Sr in a precipitate form by adding an inorganic such as carbonate to the waste LiCl salt from the electroreduction process. Another way is to simultaneously partition both Cs and Sr from a waste LiCl salt by using either Czochralski or zone freezing technologies. Finally, rare-earth elements including Y in the spent LiCl-KCl waste generated during the course of an electrorefining are removed in the form of an oxide precipitate by using an air oxidation. KAERI is focusing on a total recycling of the waste salts to each process unit after removing the fission products arising from different salts, with a minimum waste salt release to a permanent repository. This paper provides the experimental conditions to separate these fission products from each process unit and also evaluates the separation efficiency. In addition, this work will present and discuss the waste forms to be disposed of and a reference flow sheet for removing the fission products from a pyroprocess.

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* The full text of this paper was unavailable at the time of publication.

Current progress in R&D on MSR fuel cycle technology in the Czech Republic

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Abstract

The Czech R&D programme in the field of partitioning and transmutation is based on the molten salt reactor system concept with fluoride-salts-based liquid fuel, the fuel cycle of which is based on pyrochemical fluoride partitioning of spent fuel. There are three main pyrochemical partitioning techniques proposed for processing and/or reprocessing of MSR fuel: fluoride volatilisation processes, molten salt/liquid metal extraction processes and electrochemical separation processes. Two of the processes – fluoride volatility method and electrochemical separation process from fluoride media are under experimental development in the Nuclear Research Institute Řež plc.

R&D on the fluoride volatility method is focused to the development and experimental verification of a semi-pilot technology for LWR spent fuel reprocessing, which may result in a product the form and composition of which might be applicable as a starting material for the production of liquid transuranium fuel for a molten salt transmutation reactor. R&D on electrochemical separation processes from fluoride melt media is aimed mainly to the "on-line" reprocessing (partitioning) of the liquid fuel circulating in the primary (fuel) circuit of molten salt reactor. Besides the two main experimental partitioning activities, the flow sheeting research is in the focus of interest as well. The paper summarises the results achieved in the development of pyrochemical partitioning technologies mentioned above and outlines the future activities in the Czech P&T programme.

Introduction

The Czech R&D programme in the field of partitioning and transmutation is based on the molten salt reactor system concept with fluoride-salts-based liquid fuel, the fuel cycle of which is based on pyrochemical fluoride partitioning of spent fuel. The molten salt reactor (MSR) represents one of promising advanced reactor type assigned to the Gen-IV reactor systems. It can be operated either as a thorium breeder within the ^{232}Th-^{233}U fuel cycle or as an actinide burner (transmuter) incinerating transuranium fuel. Molten salt reactor incinerating transuranium fuel is often denoted "molten salt transmutation reactor" (MSTR). Essentially the main advantage of MSR (or MSTR) comes out from the prerequisite, that this reactor type should be directly connected with the "on-line" reprocessing of circulating liquid (molten salt) fuel. Besides, the on-line fuel salt clean-up is necessary over the long run to keep the reactor in operation. As a matter of principle, it permits to clear away typical reactor poisons like xenon, krypton, lanthanides, etc., and possibly also other products of burned plutonium and transmuted minor actinides. The fuel salt clean-up technology should be linked with the fresh MSR fuel processing to continuously refill the new fuel into the reactor system. On the other hand, the technologies of fresh transuranium molten salt fuel processing from the current LWR spent fuel and of the on-line reprocessing of MSR fuel represent two killing points of the whole MSR technology, which have to be successfully solved before MSR deployment in the future.

The main fuel processing and reprocessing technologies proposed for the MSTR fuel cycle are generally pyrochemical, the majority being fluoride technologies [1]. This is caused by the fact that MSTR fuel is constituted by a mixture of molten fluorides.

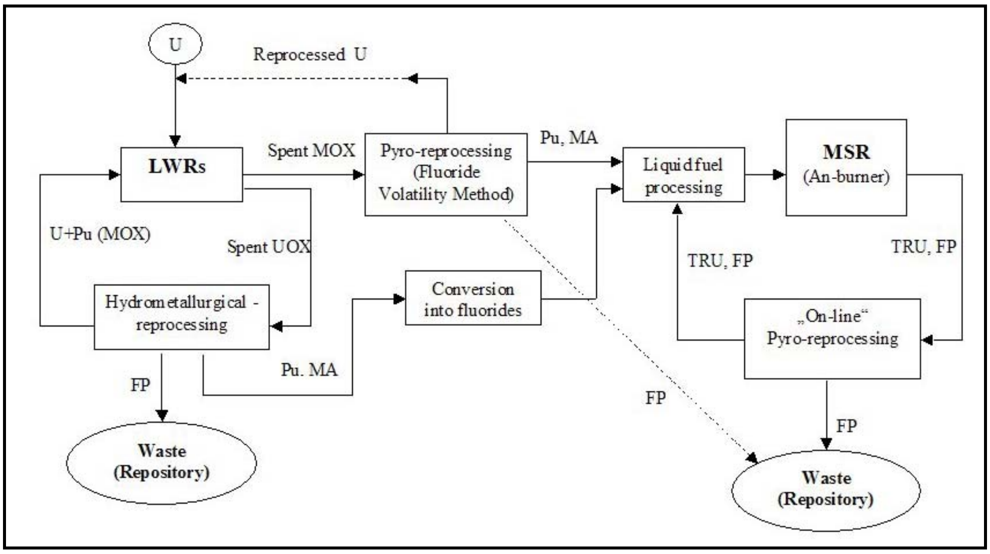
Three main pyrochemical separation techniques are proposed for processing and subsequent reprocessing of MSTR fuel:

- fluoride volatilisation processes;
- molten salt/liquid metal extraction processes;
- electrochemical separation processes.

As the fluoride volatilisation process can be profitably used as the initial step of transuranium fuel processing, the molten salt/liquid metal extraction and/or electrochemical separation processes could be used for "on-line" reprocessing of circulating MSTR fuel. The emplacement of the technologies is shown in Figure 1 describing the Czech concept of partitioning and transmutation.

Figure 1: Czech P&T concept – double-strata strategy with MSTR in second stratum

MSTR = MSR – actinide burner



There are two pyrochemical technologies convenient to the MSTR fuel cycle, which are currently under development in the Czech Republic. Both technologies are under development in the Nuclear Research Institute Řež plc. The first technology is the fluoride volatility method (FVM) designated to the reprocessing of spent oxide fuels from current or future light water reactors, the second one is electrochemical separation from fluoride molten salt media designated either to the final processing of liquid fuel for MSTR or to the "on-line" reprocessing of circulated spent MSTR fuel.

Progress in the fluoride volatility method

The fluoride volatility method (FVM) is regarded as a promising advanced pyrochemical reprocessing technology, which can be used for reprocessing mainly oxide spent fuels coming from future LWR or Gen-IV fast reactors. The technology should be chiefly suitable for the reprocessing of advanced oxide fuel types, *e.g.* fuels with inert matrixes and/or fuels of very high burn-up, high content of plutonium and very short cooling time, which can be hardly reprocessed by hydrometallurgical technologies.

FVM is based on a separation process, which comes out from the specific property of uranium, neptunium and plutonium of forming volatile hexafluorides whereas most of fission products (lanthanides) and higher transplutonium elements present in irradiated fuel form non-volatile trifluorides. This property has led to the development of several technological processes based on fluoridation of irradiated fuel either by strong fluorinating agents like BrF_3, BrF_5, ClF_3 or even by pure fluorine gas. Major former activities were carried out in US at Brookhaven, Argonne and Oak Ridge laboratories, in France at Fontenay-aux-Roses, in Belgium at Mol, in the former Soviet Union at Dimitrovgrad and in the former Czechoslovakia at Řež [2-5].

The reprocessing technology based on FVM consists of the following main operations:

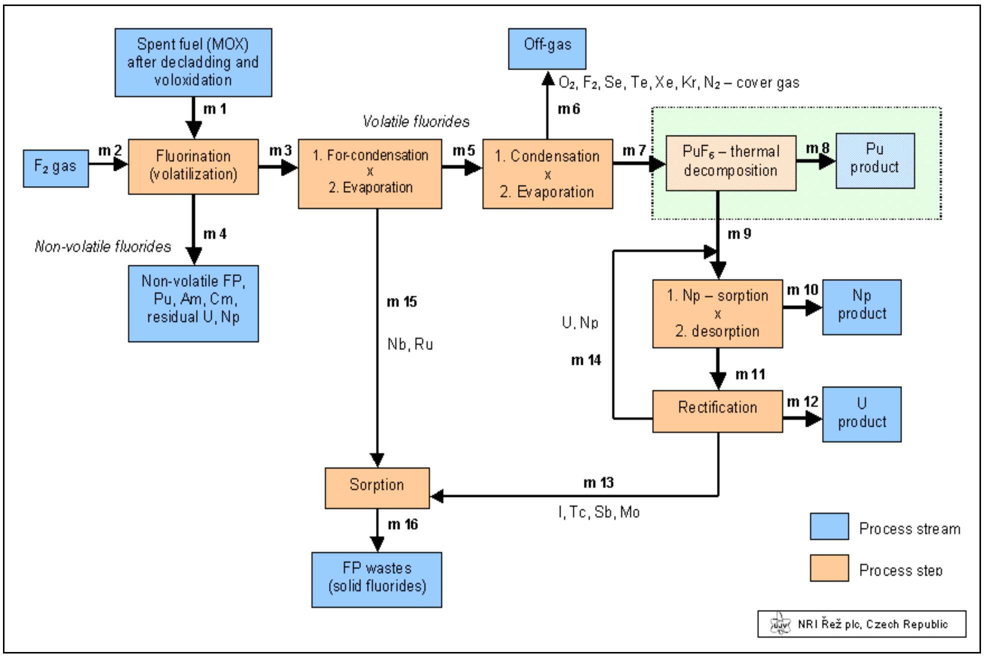
- removal of the cladding material from spent fuel elements;
- transformation of the fuel into a powder form of a granulometric composition suitable for the fluorination reaction;
- fluorination of the fuel (the purpose of this operation is the separation of the uranium component from plutonium, minor actinides and most fission products);
- purification of the products obtained.

The first two steps represent preparatory stages for FVM itself and can be realised separately from the FVM. Whereas the early fluorination techniques of the FVM were fluidised bed processes, now the direct flame fluorination of powdered fuel is considered as the most promising unit operation for future industrial application. This method of fluorination in the frame of the FVM, which was first used in 1980s, is now under development at the Nuclear Research Institute Řež plc.

Flame fluorination reaction of spent oxide fuel is a basic unit operation of the whole process. The reaction between the fuel powder and pure fluorine gas is spontaneous and highly exothermic. Usual temperature of ignition is over 250°C. Subsequently the temperature in the flame can reach in the reactor the range of 1 500-1 700°C. Based on the fluorination reaction, the main partitioning of spent fuel is realised directly in the fluorination reactor. Whereas the volatile products of the fluorination reaction leave the apparatus, the non-volatile fluorides remain caught in the fluorinator bottom in the form of ash. The further separation of most of individual components forming volatile fluorides is generally possible by sorption, condensation or distillation processes.

The Nuclear Research Institute Řež plc has been developing the FVM in the frame of national P&T project SPHINX as a "front-end" technology devoted to the fuel cycle of MSTR. Here, the present day R&D represents the follow-up of the former long-lasting R&D activities in FBR fuel reprocessing. The current experimental R&D programme of the verification pyrochemical reprocessing of current and advanced oxide fuel types was launched in 2004. The process flow sheet of investigated technology is based on the direct fluorination of powdered spent fuel by fluorine gas and on the subsequent purification of volatile products by condensation, sorption, thermal decomposition and distillation (Figure 2).

Figure 2: Process flow sheet of fluoride volatility method



The current experimental R&D programme is focused mainly on the technological verification of individual unit operations, apparatuses and material research and to the process control. The experimental semi-technological line called FERDA, shown in Figure 3, was manufactured and placed in the alpha-radiochemical laboratory of the NRI Řež plc.

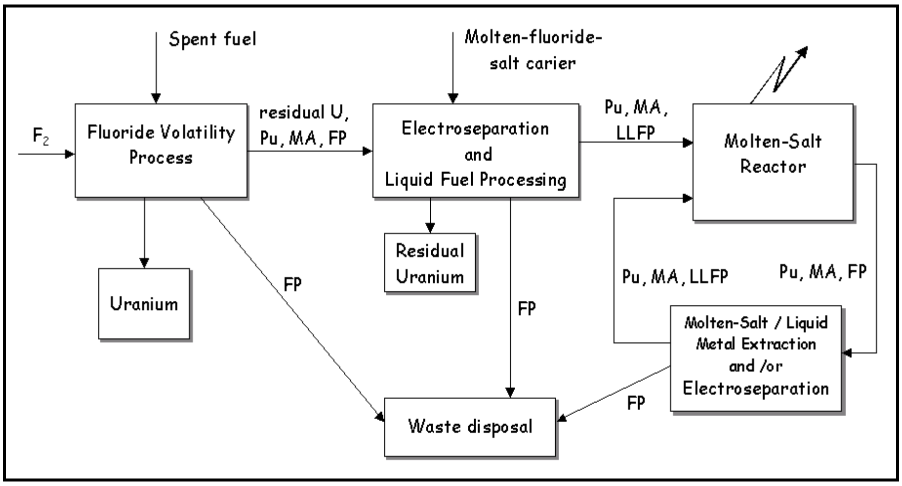
Figure 3: Experimental line FERDA for R&D on FVM

The main present experimental effort is focused on the mastering of the fluorination process and the elimination of the bottlenecks of the technology. After the tests done with the uranium fuel, the present programme is focused on the verification of the main unit operations with the simulated spent oxide fuel constituted from a mixture of uranium oxides and non-radioactive oxides representing selected fission products (lanthanides, Cs, Sr, etc.). The next series of experiments should verify the suitability of the technology for reprocessing of oxide fuels with inert matrixes.

FVM has a good potential to be used within the fuel cycles of several current or advanced reactor types for reprocessing of current or future oxide fuel types. The main attractiveness to use FVM as the “front-end” technology of MSTR is due to the conversion of the oxide form of the original spent fuel into fluorides – the chemical form of molten salt reactors. Here the main aim of the process is to remove uranium and some part of fission product from the LWR spent fuel and to prepare a rare transuranium fuel in the form of fluorides for final preparation of TRU molten salt fuel.

As the FVM is not able to separate trivalent minor actinides (Am, Cm) from most fission products, the final processing of TRU molten salt fuel has to be done by some subsequent technology. Either molten salt/liquid metal reductive extraction or electrochemical separations are generally proposed as suitable methods. The use of a combination of FVM with other pyrochemical processes within the MSTR fuel cycle is shown in Figure 4.

Figure 4: Fuel cycle of molten salt transmutation reactor



Progress in electrochemical separations from fluoride molten salt media

The use of electrochemical separation processes seems to be promising as for final TRU fuel processing for MSTR (An-burner) as for on-line reprocessing technology of MSR. Here the principle of the partitioning is similar as for MSR-Th-breeder as for MSTR.

The development of on-line reprocessing (fuel salt clean-up) technology represents a very specific problem affecting even the MSR design, reactor core chemistry and a choice of structural materials. Particularly the link to reactor core chemistry is close because the chemical reaction rate and their character in the reactor have to be compensated by the reprocessing technology. Special attention should be paid to the selection of carrier molten salt, which must exhibit several basic properties (*e.g.* good thermal conductivity, appropriate melting point, low vapour pressure, radiation stability, sufficient solubility of actinides and last but not least the reprocessability by adequate separation techniques). Based on these requirements, the ^{7}LiF-BeF$_2$ eutectics remain the basic carrier salt candidate among several others, sometimes considered or proposed molten salt mixtures, for this purpose [6].

The carrier molten salt of primary (fuel) circuit of MSTR is proposed to be similar to MSR breeder. An addition of NaF into the original ^{7}LiF-BeF$_2$ eutectic (FLIBE) is recommended to improve the solubility of trifluorides of plutonium, americium and curium, which is low in FLIBE. As the thermodynamical and electrochemical stability of BeF$_2$ emerged to be insufficient (the carrier must be more stable then separated constituents) for partitioning processes, other molten fluoride salt mixtures were chosen as carriers for individual steps of separation technology. Generally, only fluoride molten salts have been considered as carrier components for reprocessing technology to avoid possible contamination of MSTR primary circuit by other anions (*e.g.* chlorides) from the separation processes.

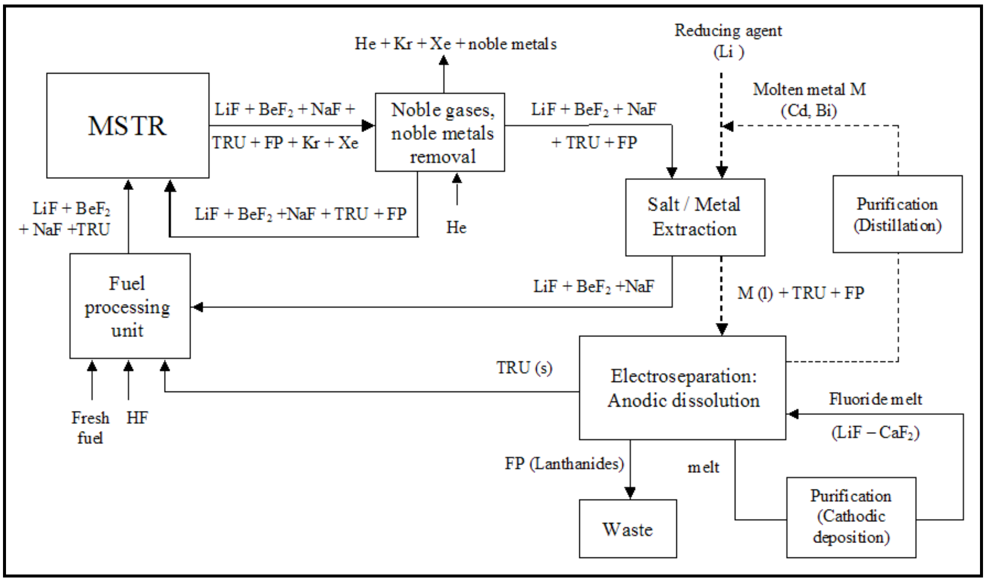
As the molten salt/liquid metal extraction processes dedicated to actinide/lanthanide separation were intensively studied during the MSRE and MSBR projects, the electrochemical separation represents the other possible and promising partitioning technique suitable for on-line reprocessing of MSR (MSTR) spent fuel. The electrochemical methods for separation actinides and fission products from selected molten fluoride salt carriers are under development in the NRI Řež plc. The typical molten salt carriers under study are: LiF-NaF-KF eutectic (often called FLINAK), LiF-CaF_2 eutectic and in LiF-BeF_2 eutectic (FLIBE).

Based on the electrochemical measurements, there seems to be a significant chance that two preliminary electrochemical techniques will be proposed – a cathodic deposition method and an anodic dissolution (oxidation) method as the main partitioning processes for separation of actinides from lanthanides within the MSR (MSTR) spent fuel on-line reprocessing. The more detailed study of electrochemical separation processes including the latest results achieved are described in a special paper of K. Chuchvalcová Bímová, *et al.* [7].

The general spirit of MSR (MSTR) on-line reprocessing is to keep the reactor in steady-state conditions by continuous cleaning-up of the primary (fuel) circuit salt. It means, that some part of the salt circulating in the primary circuit is piped to the reprocessing unit, where the fission products are extracted and then moved to waste, whereas the separated actinides are dissolved again in the carrier salt and returned back into the primary circuit. As there is all the time the same concentration of fission products in the primary circuit, the removal of these elements in reprocessing unit need not be absolute, however no actinides can be moved into the waste stream. A conceptual flow sheet of MSTR on-line reprocessing technology is drafted in Figure 5.

Figure 5: Conceptual flow sheet of MSTR – SPHINX on-line reprocessing technology

TRU – transuranium elements, FP – fission products



The reprocessing technology is based on primary non-selective molten-salt/liquid metal reductive extraction and subsequent selective electrochemical separation processes. Li and molten Bi or Cd are proposed to be used as reduction and extraction agents, respectively.

The reason for such a combination of salt/metal extraction and electroseparation resides in fact that the lanthanides should be removed prior to the actinides, which should remain in the main fuel stream and go back to the molten salt reactor. According to the experimentally measured thermodynamic properties of selected lanthanides and actinides, fluorides of the lanthanides are more stable and can be reduced neither electrochemically nor chemically prior to actinides without simultaneous reduction of the latter ones. The proposed flow sheet is based on the removal of all elements from the carrier salt in a form of metallic mixture, from which it would be possible to selectively remove only lanthanides using the electrochemical anodic dissolution method.

Conclusion

Successful solution of the MSR fuel cycle technology development seems to be one of the crucial steps before industrial deployment of MSR systems. As the MSR (MSTR) processing/reprocessing technology must meet special demands (like radiation resistance, compactness, exclusion of moderating agents, compatibility with the carrier molten salt type and with the structural material of MSR primary circuit, acceptable process reaction rate and process workability by remote handling), the pyrochemical separation processes seems to be the only technologies which can be generally applied. The fluoride volatility method has a good potential to be used as the first step for fluoride TRU fuel processing and the electrochemical separation technologies could be particularly used for the MSR fuel on-line reprocessing.

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Effects of repository conditions on environmental impact reduction by recycling

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Abstract

The environmental impacts (EI) of high-level wastes (HLW) disposed of in a water-saturated repository (WSR) and in the Yucca Mountain Repository (YMR) for various fuel cycle cases have been evaluated and compared to observe the difference in the recycling effects for differing repository conditions. With the impacts of direct spent fuel disposal in each repository as the reference level, separation of actinides by UREX+ and borosilicate vitrification clearly reduces the environmental impacts of YMR, while separation by PUREX and borosilicate vitrification would not necessarily reduce the environmental impact of WSR.

Introduction

The objectives of this study are to evaluate the environmental impact (EI) of high-level wastes (HLW) disposed of in a geologic repository for various fuel cycle cases, and to observe the difference in the recycling effects for differing repository conditions based on the previous studies for waste conditioning [1], radionuclide transport [2] and their joint application for fuel-cycle evaluations [3,4]. This study compares a water-saturated repository with the Yucca Mountain Repository (YMR). The EI [2] refers to the total toxicity index of radionuclides existing in the region exterior to the repository after waste canister failure.

Models and cases for calculations

For each case, the computation has been done as follows. First, the spent fuel composition after irradiation, reprocessing and cooling before vitrification is determined by ORIGEN 2.1 [5]. Second, the composition and the number of canisters for vitrified HLW [1] are determined. Third, the total EI of radionuclides existing in the environment, released after package failure [2] as a function of time is evaluated, and the peak EI (PEI) has been found. Lastly, the PEI per electricity generation (EIE) has been computed by dividing the PEI by the total electricity generated.

For the water-saturated repository, the EIE resulting from disposal of: (1) PWR-UO_2 spent fuel, (2) HLW from PWR-UO_2 fuel [7], (3) HLW from PWR-MOX fuel [8] and (4) HLW from fast reactor fuels [9] have been evaluated. For detailed parameter values, refer to [3]. For YMR, (5) light-water reactor (LWR) spent fuel, and (6) HLW from LWR-UO_2 fuel by UREX+ with various different separation efficiencies are evaluated. Detailed parameter values for this case are reported in [2,4]. Table 1 summarises the main results of these cases.

Water-saturated repository combined with PUREX

Burn-up, PUREX and vitrification

The high-level liquid waste (HLLW) inventory after cooling and reprocessing is calculated via ORIGEN 2.1. Initial fuel composition and input data for ORIGEN 2.1 calculation are given in Table 1(a). A PUREX process is assumed for the PWR fuel. The waste conditioning and radionuclide models are here to link the HLLW inventory to the environmental impact resulting from their disposal. Resulting from the solvent clean-up during reprocessing, the addition of Na to the HLLW is assumed [10]. This is important because one of the constraints for HLLW loading in borosilicate glass is related with the mass of Na in the HLLW [see below in this section and in Table 1(a)].

For FR fuels, an aqueous process similar to PUREX for separation of minor actinides (Np, Am and Cm) is assumed as well as for U and Pu with an overall actinide separation efficiency of 99.5%. This approach has not been retained for the PWR-MOX case due to the limited recyclability of its spent fuel [11]. The following elements are not retained in the waste stream because they are volatile: H, He, C, Ne, Cl, Ar, Kr, I, Xe and Rn. It is considered that the equilibrium mass flow of FR back-end fuel cycle is weakly sensitive to separation efficiency higher than 99%. We assume that the corrosion products (Fe, Ni and Cr) in the HLLW will not be included in the vitrified HLW.

HLLW generated from reprocessing is assumed to be solidified in borosilicate glass, contained in a metal canister (see Table 2 for its dimensions), and cooled down in an interim storage before geologic disposal. In order to ensure the feasibility of this step, the mass loading of radionuclides in a canister needs to comply with several constraints. In this study the specifications and constraints summarised in Table 2 are assumed. The waste conditioning module developed at UCB [1] calculates the composition of vitrified HLW in a canister in such a way that the waste radionuclide mass loading is maximised while satisfying a given set of constraints [7]. This optimisation is done through a linear programming approach. The two main outputs are the composition vector of vitrified HLW in the canister and the resulting number of canisters per MTHM of spent fuel reprocessed.

For Pu-rich fuels like PWR-MOX or FR fuels, a substantial cooling time before reprocessing (T_b) has been chosen. The relatively high concentrations of strong alpha-emitters like Cm or Pu isotopes within spent fuels may damage the organic solvent TBP and hence prevent an efficient reprocessing.

Table 1(a): Parameter values for the cases for water-saturated repository

		PWR UO_2	PWR MOX	FR (core/axial)
Burn-up conditions		**Cases (1, 2)**	**Case (3)**	**Case (4)**
Fuel composition before irradiation (g/MTHM)				
^{234}U		450	0	0
^{235}U		45 000	1 856	1 722/833
^{236}U		250	0	0
^{238}U		954 300	926 144	571 583/276 942
^{238}Pu		0	1 224	1 637
^{239}Pu		0	40 608	80 568
^{240}Pu		0	16 632	47 798
^{241}Pu		0	8 064	6 404
^{242}Pu		0	4 248	5 812
^{237}Np		0	0	744
^{241}Am		0	1 224	2 981
^{243}Am		0	0	1 488
^{244}Cm		0	0	1 488
ORIGEN cross-section library numbers		604/605/606	210/211/212	311/312/313 (core); 314/315/316 (blanket)
Thermal output (MW/MTHM)		38	37.7	35.9
Operating days (EFPD)		1 184	1 592	3 200
Discharged burn-up/core average (GWd/MTHM)		45	60	115/150
Power allotment (core/axial blanket, %)		–	–	94.4/5.6
Capacity factor, C_{factor}		0.9		0.8
Conversion efficiency, C_{eff}		0.33		0.42
PUREX conditions		**Case (2)**	**Case (3)**	**Case (4)**
Cooling time before reprocessing, T_b (yr)		3	10	7
Cooling time between reprocessing and vitrification, T_a (yr)		1	1	1
Fractions removed from HLLW by PUREX (%)	U, Pu	99.5	99.5	99.5
	Np, Am, Cm	0	0	99.5
	I	99	99	99
	H, C, Cl, He, Ne, Ar, Kr, Xe, Rn	100		
Vitrification results				
Mass of waste oxides in a canister (kg/can)		74	47	84
Mass of borosilicate glass matrix in a canister (kg/can)		314	325	307
Number of canisters per MTHM of fuel, N_{can} (can/MTHM)		1.27	2.00	1.97

Table 1(b): Parameter values for the cases of UREX1a+ combined with Yucca Mountain Repository

Burn-up conditions	**Cases (5) (6-1) (6-2) (6-3)**		
Fuel composition before irradiation (g/MTHM)			
^{235}U	43 000		
^{238}U	957 000		
ORIGEN cross-section library numbers	219/220/221		
Thermal output (MW/MTHM)	40		
Operating days (EFPD)	1 250		
Discharged burn-up (GWd/MTHM)	50		
Capacity factor, C_{factor}	0.9		
Conversion efficiency, C_{eff}	0.33		
UREX1a+ conditions			
Cooling time before UREX1a+, T_b (yr)	15		
Cooling time between UREX1a+ and vitrification, T_a (yr)	0		
Fractions removed from HLLW by UREX1a+ (%)	**Case (6-1)**	**Case (6-2)**	**Case (6-3)**
U, Pu, Np, Am, Cm, Tc, Cs, Sr	95	99	99.5
H, C, I, Cl, He, Ne, Ar, Kr, Xe, Rn	100		
Vitrification results			
Mass of waste oxides in a canister* (kg/can)	513	454	407
Mass of glass matrix in a canister* (kg/can)	1 520	1 579	1 626
Number of packages** for HLW generated by UREX1a+ processing of 63 000 MTHM of fuel	2 994	2 324	2 324

* The canister dimensions are shown in Table 2.

** Five canisters are contained in one package with the dimensions of the Co disposal package shown in Table 3. Note that the total number of packages of 63 000 MTHM CSNF, *i.e.* for Case (5), is 7 886 [2,13].

Table 2: Vitrification constraints assumed

Specifications/constraints	Water-saturated	YMR
Canister height (m)	1.34	3
Canister outer radius (m)	0.215	0.305
Canister thickness (m)	0.006	0.01
Canister volume, V_c (m^3)	0.15	0.82
Empty canister weight (kg)	100	467
Total mass of a package (kg)	< 500	< 2 500
Mass fraction of Na_2O (wt.%)	< 10	< 10
Mass fraction of MoO_3 (wt.%)	< 2	< 2
Concentration of Pu (kg/m^3)	< 2.5	< 2.5
Heat emission (kW/canister)	< 2.3	–
Maximum temperature in glass (°C)	–	< 400
Volume of vitrified HLW (m^3/canister)	$< V_c$	$0.8V_c < V < V_c$

Following Ref. [10], reasonable T_b for the reference FR and PWR-MOX case are respectively of 7 yr and 10 yr. The cooling time (T_a) between reprocessing and vitrification is assumed to be 1 yr. This choice was motivated by the relative difficulty to handle and conserve HLLW compared to its solid form after discharge. Hence the allocation of cooling time before vitrification will be done by default in favour of T_b instead of T_a. Pyroprocessing, which is not considered in the present study, remains less sensitive to strong alpha-emitters than aqueous processes. Its implementation allows for a reduction of T_b; future studies will be carried out to develop a pyroprocessing model.

Water-saturated repository conditions

We consider a hypothetical geologic repository in a water-saturated geologic formation, such as those proposed in Europe, Japan and so on for these fuel cycle cases. It is assumed that each waste canister determined in the previous steps is surrounded by a system of engineered barriers, such as a metal container and a backfill. The backfill, such as bentonite, around the waste canister is assumed to have low water permeability, resulting in diffusion-dominant radionuclide transport in the engineered barriers. The geochemical environment is assumed to be reducing, resulting in low solubilities for actinides. Table 3 summarises and compares the repository conditions for the water-saturated repository [12] and for the Yucca Mountain Repository [2]. The UO_2 matrix (and/or slightly oxidised, yet iso-structural alternation phases such as U_4O_9 or U_3O_7) of spent fuel is considered thermodynamically stable under suitably reducing conditions. Thermodynamic stability is never true for borosilicate glass as it is an intrinsically thermodynamically unstable phase.

Uncertainty associated with repository conditions might induce a substantial variability of the canister failure time (CFT). The assumed reference value for CFT is 10 000 yr. Following the radionuclide release from a failed canister, PEI is computed via the code previously developed [2]. In the present study, interactions among canisters are neglected. Each canister is separated with sufficiently large distances, and acts as an independent source. With this independent canister assumption, PEI is linearly proportional to the total number of canisters and consequently RIE is neither dependent on the number of canisters nor on the canister spatial disposition.

Yucca Mountain Repository combined with UREX

UREX1a+ and vitrification

In this study, while the Yucca Mountain Repository is designed to contain both commercial spent nuclear fuel (CSNF) and defence HLW and spent fuels including naval spent fuels, we have focused on 63 000 MT of CSNF. We compare the EI from direct disposal of CSNF with that from the HLW after UREX+1a separation, with the assumption that the resulting HLW is vitrified in a borosilicate glass matrix. The analysis has started with the determination of CSNF compositions. While the CSNF compositions are partially reported in the *Final Environmental Impact Statement Report* [13], the complete inventory of spent fuel including all minor fission product isotopes has been obtained by using ORIGEN 2.1, and compared with the compositions in [13]. The same burn-up conditions as given in Ref. [13] have been used. See Table 1(b).

Table 3: Repository conditions assumed

Parameters		Water-saturated [12]	YMR[2]
Canister/package failure time, T_f (yr)		10 000	75 000
Radius of waste package (m)		0.21	See note
Length of waste package (m)		1.34	
Pore velocity of groundwater in surrounding rock (m/yr)		1	0.77
Porosity of the surrounding medium		10%	10%
Diffusion coefficient in the surrounding medium (m^2/yr)		3E-2	3E-2
Solubility in groundwater (mol/m^3)	Se	3.0E-06	1.0E+02
	Zr	1.0E-03	6.8E-07
	Nb	1.0E-01	1.0E-04
	Tc	4.0E-05	High
	Pd	1.0E-06	9.4E-01
	Sn	1.0E-03	5.0E-05
	Cs	High	High
	I	High	High
	Sm	2.0E-04	1.9E+02
	Pb	2.0E-03	1.0E-02
	Ra	1.0E-09	2.3E-03
	Ac	2.0E-04	1.9E+02
	Th	5.0E-03	1.0E-02
	Pa	2.0E-05	1.0E-02
	U	8.0E-06	4.0E-01
	Np	2.0E-05	1.6E+01
	Pu	3.0E-05	2.0E-01
	Am	2.0E-04	1.9E+02
	Cm	2.0E-04	1.9E+02
	Si	0.21	2.1

The radius and the length of the CSNF package are assumed 0.792 m and 5.165 m, respectively. Those for the Co disposal package are 0.60 m and 5.217 m, respectively.

After discharge from the reactor, the spent fuel is left to cool for a time T_b, after which it undergoes separation by the UREX+1a scheme. This scheme extracts U, Tc, Cs/Sr and TRU with a given separation efficiency into different streams. We consider three values: 95%, 99% and 99.5%. All other non-volatile fission products are assumed to be left in the high-level liquid waste stream. After processing, the HLLW is left to cool some additional amount of time T_a before it is vitrified.

The same waste conditioning module [1] was used to calculate the composition of vitrified HLW in a canister with the dimensions and the vitrification constraints given in Table 2. For final disposal, five of these vitrified HLW canisters are assumed to be contained in a single waste package, which has the same dimensions as those of the Co disposal waste package (see Table 3).

The waste package would be stored in an interim storage before it is placed in the Yucca Mountain Repository, to meet the heat emission requirement. The maximum initial heat power from a waste package in YMR is limited at 11.8 kW per package [14]. With the maximally-loaded vitrified HLW obtained in this study, where Cs and Sr are also assumed to be removed, the heat emission at the time of vitrification is calculated to be approximately 3 kW/package, in comparison with 25.5 kW/package with all Cs and Sr isotopes retained in the HLW [1]. Therefore, interim storage for the purpose of cooling to meet the YMR heat emission requirement would not be necessary.

The parametric investigations [4] observed that waiting for a total of 15 years to let the spent fuel or HLW cool, the removal of the heat-emitting fission products Cs and Sr does not allow for a larger waste mass loading. This is because the temperature constraint is not an active constraint in the linear programming model after 15 years. It is the Mo mass fraction constraint that becomes the governing constraint, indicating that removal of heat-emitting radionuclides may not necessarily have a significant effect on waste loading as expected. In this study, as Table 1(b) shows, we have chosen the cooling time value of 15 years before and no cooling after partitioning. Considering that many spent fuel assemblies have already been stored, this 15-year cooling time assumption is reasonable, and also matches with the assumption made in [13].

Yucca Mountain Repository conditions

Table 3 summarises and compares the repository conditions for the water-saturated repository [12] and for the Yucca Mountain Repository [2]. The geochemical environment in YMR is assumed to be oxidising, resulting in relatively high solubilities especially for actinides. Also, in the oxidising environment, the uranium oxide as well as the borosilicate glass is considered thermodynamically unstable. In Ref. [2], it was pointed out that the dissolution times of the borosilicate glass in a Co disposal package (approximately 7 500 kg) and the uranium oxides in a CSNF package (approximately 8 000 kg) are in the same range, *i.e.* 400 000 years after package failure. The assumed reference value for CFT is 75 000 yr. Following the radionuclide release from a failed canister, PEI is computed via the code developed at UCB [2].

Results

Water-saturated repository case

The end results for the burn-up, separation processes, and vitrification are the number of vitrified HLW canisters/packages, summarised in Table 1. The results for the water-saturated repository case with Pu recycle with MOX fuel in PWR and FR, shown in Table 1(a), indicate that the Pu recycle results in greater numbers of canisters per MTHM of spent fuel. Case (2) (HLW from UO_2 spent fuel reprocessing) generates 1.27 cans, while approximately 2 cans are generated from Case (3) (PWR-MOX) and Case (4) (FR). Table 1(a) also shows electricity generation supported by a 10 000 canister repository (EGR) for Case (2) (130 TWd), Case (3) (110 TWd) and Case (4) (310 TWd).

The irradiation of MOX fuel with high burn-up leads to a significant build-up of ^{244}Cm that is not removed by the PUREX process assumed for Cases (2) and (3). Furthermore, the decrease of the actinide heat emission following Cm decay is balanced by the ^{241}Am build-up via ^{241}Pu (14 yr half-life). This high level of heat emission for the PWR-MOX HLLW is responsible for a substantial dilution of waste inside the borosilicate glass and a consecutive reduction of the EGR. Greater EGR for FR cases are related to their high burn-up and the low heat emitting HLLW resulting from the Cm and Am removal. Another benefiting effect is obtained from the higher conversion efficiency assumed for FR.

Figures 1-4 show the results of the environmental impact assessment for the water-saturated repository for Cases (1) to (4). In Table 4, the peak values of the EIE are summarised. In Case (2) (PWR-UO_2), the peak value of the EIE is the combination of two main radionuclides: ^{243}Am and ^{239}Pu. Compared to Case (3) (PWR-MOX), Case (2) has the smaller Pu isotope concentrations. The EIE for Case (4) is one order of magnitude smaller than that for Case (3).

If compared to its PWR-UO_2 counterpart, *i.e.* Case (2), the direct disposal [Case (1)] leads to a significantly lower EIE (see Figure 1.) The EIE is 1.7E7m^3/GWyr which remains lower than the EIE for the FR case by two orders of magnitudes. This lower EIE for the direct disposal case is explained by the assumed low solubility of the uranium matrix in water (8.0E-6 mol/m^3), 4 orders of magnitude smaller than that of silica. From an environmental impact point of view, this indicates that a direct disposal scenario is preferable to any other types of scenarios based on reprocessing and use of borosilicate glass, if the repository chosen implies a low solubility of the uranium matrix in comparison to the borosilicate glass matrix.

Yucca Mountain Repository case

The results for the YMR case, shown in Table 1(b), indicate that the number of waste packages would be decreased to approximately 1/3 of that (7 886) of original CSNF packages. With 15 years of total cooling time for the cases of 99% and 99.9% separation efficiency, the number of waste packages stays the same. For the case of 95% efficiency, the waste package number is increased by about 600, due to the Pu constraint becoming active.

Figure 5 shows the results of the environmental impact assessment for the YMR for Case (5), direct disposal of UO_2 spent fuel. The results for total EIE for Cases (6-1) to (6-3) with the three different separation efficiencies in the UREX1a+ process are directly proportional to the separation efficiency. In Table 4, the peak values of the EIE are summarised.

Table 4: Environmental impact per electricity generation (EIE) (m^3/GWyr)

Water-saturated repository	**(1) PWR UO_2 spent fuel**	**Vitrified HLW from PUREX**		
		(2) from PWR UO_2 with 99.5% removal for U and Pu	**(3) from PWR MOX with 99.5% removal for U and Pu**	**(4) from FR with 99.5% removal for all actinides**
EIE (m^3/GWyr)	1.7E7	1.4E9	7.5E9	8.3E8
Relative to spent fuel direct disposal [Case (1)]	1	82	440	49

Yucca Mountain Repository	**(5) LWR UO_2 spent fuel**	**Vitrified HLW from LWR UO_2 by UREX+**		
		(6-1) 95% removal for all actinides	**(6-2) 99% removal for all actinides**	**(6-3) 99.5% removal for all actinides**
EIE (m^3/GWyr)	4.9E9	1.2E9	2.3E8	1.2E8
Relative to spent fuel direct disposal [Case (5)]	1	0.24	0.047	0.024

Figure 1: Environmental impact per electricity generation (in m^3 of water per GWyr) for the direct disposal of PWR-UO_2 Case (1)

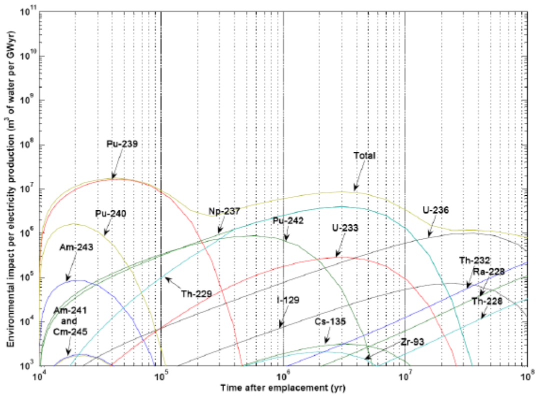


Figure 2: Environmental impact per electricity generation (in m^3 of water per GWyr) for Case (2) (PWR-UO_2)

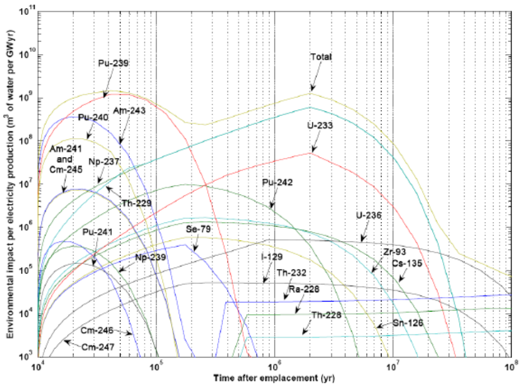


Figure 3: Environmental impact per electricity generation (in m^3 of water per GWyr) for Case (3) (PWR-MOX)

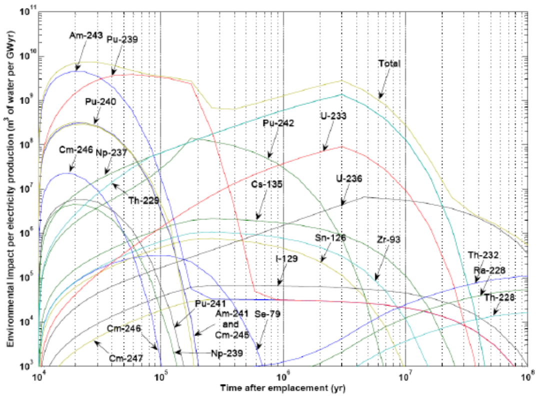


Figure 4: Environmental impact per electricity generation (in m^3 of water per GWyr) for Case (4) (FR)

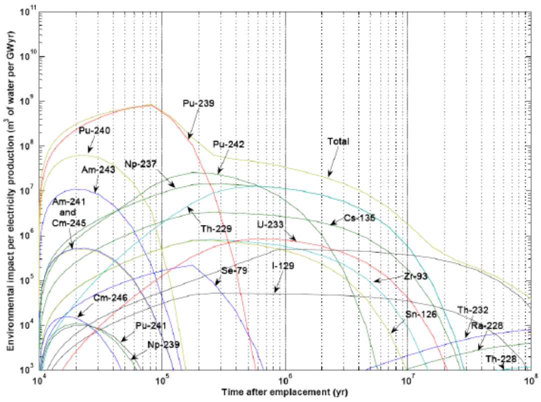
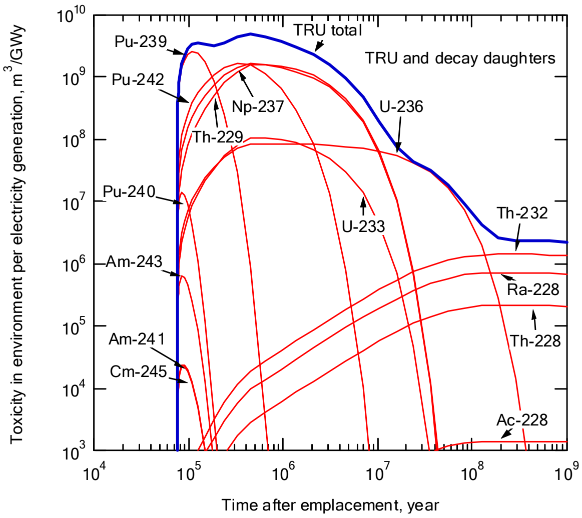


Figure 5: Environmental impact per electricity generation (in m^3 of water per GWyr) for Case (5) for YMR



Discussions

A comparison of Figures 1 and 5 shows that the EIE for the water-saturated repository is more than two orders of magnitude smaller than that for YMR. This is considered to result from the difference in assumed U solubilities for both repositories. In this study, the canister failure time has been assumed to be relatively short (10 000 years for the water-saturated repository and 75 000 years for YMR). The package lifetime could be significantly longer, according to recent studies, some of which indicate 1 million year lifetime [15]. With nearly two orders of magnitude longer package lifetime, most Pu isotopes would have decayed out in the package before failure. A parametric calculation with a 1 million year CFT instead of 75 000 years for Case (5) shows 3.0E9 m^3/GWyr, which is about 60% lower than the value shown in Table 4.

A comparison of the EIE of Case (4) with that of Case (6-3) shows that, with the same level of separation efficiencies, the EIE of both repositories become similar to each other. This results from the facts that the waste matrix is assumed to be borosilicate glass, and that the mass loading of hardly soluble actinide elements in a waste canister/package is so low due to 99.5% separation efficiency that they are released congruently with borosilicate glass dissolution, not limited by their solubilities. Note, however, that Case (6-3) does not reflect the effects of recycle of separated actinides in any fuel cycle. Only separation of actinides and some FP with UREX1a+ have been assumed. With determination of the recycle system after UREX1a+ separation, we can determine more realistic HLW compositions for disposal. The previous study [16] is one of such efforts, where it was shown that by burning recovered actinides in accelerator-driven transmuters an additional 500 GWyr is generated while generating different types of HLW.

While direct comparison of EIE values between two repositories is not so meaningful without uncertainty information, we can still make a comparison in terms of the effects of reprocessing and vitrification on repository impact. For the assumed water-saturated repository, reprocessing and vitrification could make the EIE significantly greater because thermodynamically stable UO_2 matrix is replaced with unstable borosilicate glass while keeping toxic minor actinides in HLW. UREX+ processing and vitrification, however, could reduce the EIE of YMR because both UO_2 and borosilicate glass are unstable in the oxidising environment, resulting in similar matrix dissolution times, and because toxic minor actinides are assumed to be removed together. Thus, the effects of fuel cycle application on environmental impact reduction are significantly different for differing repository conditions.

Concluding remarks

The effects of conditions related to fuel cycles and geologic repositories on the environmental impacts from HLW geologic disposal have been compared. For comparison, direct disposal of PWR spent fuel in a hypothetical water-saturated repository and in Yucca Mountain Repository, vitrified HLW from PUREX reprocessing of PWR-UO_2, PWR-MOX and FR-MOX in the water-saturated repository, and vitrified HLW from UREX1a+ separation process of PWR-UO_2 fuel in YMR have been considered. The following observations have been made:

- Depending on repository conditions, the level of the environmental impact normalised by electricity generation would be significantly different. UO_2 spent fuel in the Yucca Mountain Repository would cause a greater impact per electricity generation basis than a repository in water-saturated conditions, where reducing environments are assumed. In such environments, uranium oxide is considered to be thermodynamically stable and solubilities of actinides are likely to be significantly smaller than those in YMR conditions.
- The environmental impacts per GWyr from vitrified HLW after removal of TRU elements would be similar between both repository conditions. This is considered to result from the facts that borosilicate glass dissolves in a similar rate in either reducing or oxidising environments due to thermodynamically unstable amorphous structure, and that the radionuclides are released congruently with matrix dissolution.
- With the water-saturated repository, changing the waste matrix from UO_2 to borosilicate glass may increase the environmental impact even if TRU isotopes are removed from HLW with high separation efficiencies. On the other hand, with the YMR, the effects of separation efficiencies appear proportionally on the environmental impact.

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Session IV

Progress in materials, including spallation targets and coolants

Chairs: C. Fazio, S. Monti

Progress in materials, including spallation targets and coolants

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Abstract

A summary on R&D activities performed in Europe, partly with the support of the European Commission, to study materials for transmutation systems and their associated technologies is given. Emphasis has been placed on structural materials selection/development, their assessment under representative conditions for spallation target and core, and the impact of the coolant technology on the materials' performance.

1 Introduction

The objective of partitioning and transmutation (P&T) of high-level nuclear waste is the reduction of the burden on geological storage in terms of waste mass minimisation, heat load and the source of potential radiotoxicity.

Innovative fast neutron reactors cooled with Na, Pb, He and accelerator-driven systems [cooled with lead (Pb) or lead-bismuth eutectic (LBE)] show the best transmutation performance. However, the preliminary design and safety requirements for these systems imply challenging issues on materials such as compatibility with different coolants, their application over a range of different operating temperatures, and high irradiation dose due to the objective to reach high burn-ups.

The importance of material-related issues and challenges has been recognised in the framework of international initiatives such as Generation-IV and within most national programmes devoted to the development of innovative fast reactors. In Europe, the European Commission has promoted, over the last ten years, projects on materials performance assessment, in particular to support the development of reactor systems and neutron spallation targets cooled with heavy liquid metals (Pb and LBE). As reference materials the ferritic/martensitic steel T91 and the austenitic steel AISI316L have been selected and tested for their use as structural materials for the neutron spallation target MEGAPIE (1 MEGAwatt Spallation Target PIlot Experiment) and for accelerator-driven systems (ADS). On the other hand, in the frame of critical fast reactor system development, where higher temperatures (> 500°C) and higher burn-ups (≥ 20%) have been envisaged, alternative materials such as the Oxide Dispersion Strengthened (ODS) alloys have been indicated to be most promising.

In the present work, to put in perspective the material-related activities, the preferred systems, objectives and strategies to implement P&T will first be recalled, and successively past and present experimental programmes together with some significant results on the structural materials selected for their use in fast neutron spectrum systems and neutron spallation targets will be summarised.

2 Transmutation performances

The management of the spent fuel from nuclear power plants is different according to the different perspectives and scenarios in different countries. However common features to the different options are the need of a deep geological repository, the requirement to reduce the burden of the radiotoxic elements to be disposed in it, non-proliferation issues, etc. In this context, partitioning and transmutation (P&T) is considered as a way of reducing the burden on a geological disposal [1].

Different reactor systems (fast and thermal neutron spectra) are under consideration to transmute nuclear waste. As far as the transmutation performances of these systems are concerned, this can be compared on the basis of transmutation physics characteristics such as the fission/absorption cross-section ratios for actinides, the build-up of higher actinides and the neutron balance.

The fission/absorption cross-section ratios for actinides are consistently higher for fast neutron spectrum with respect to thermal neutron systems. Thus, in a fast spectrum, actinides are preferentially fissioned, not transmuted into higher actinides. Therefore, fast neutron systems are more “efficient” in destroying actinides because fewer neutrons are lost to capture reactions before eventual fission [2]. The higher actinides’ (americium, curium, etc.) build-up within light water reactor (LWR) recycle is considerable and, due to their radioactive features, tends to be more problematic for fuel handling and fabrication in a closed fuel cycle. The comparison of the neutron balance in fast and thermal neutron systems, by evaluating the feasibility of transmutation of the different isotopes in each reactor concept, showed that in general, all of the fast reactor systems (harder neutron spectrum) exhibit a significantly more favourable neutron balance compared to the thermal systems [2]. This implies that the use of transuranics (TRU) fuel in LWR requires the addition of fissile isotopes (an economic penalty) and that the amount of minor actinides (MA) should be kept low, reducing the transmutation performance.

The transmutation performance has also been assessed by evaluating the different fuel types and coolant technologies that can be used for fast neutron (critical systems and subcritical ADS) [3]. At present, Na, Pb, He are considered for critical systems and lead (Pb) or lead-bismuth eutectic (LBE) are the reference coolant options for accelerator-driven systems [4]. Concerning the coolant options, it has been concluded that the effect of coolant choice in fast neutron system (either critical or subcritical) design will be relatively limited from the viewpoint of the MA transmutation [3].

Moreover, these studies have shown that MA transmutation effectiveness for oxide, nitride and metal fuels is comparable within few percentages.

3 P&T objectives and implementation [5]

The P&T approach has been developed within radioactive waste management strategy studies in terms of reduction of potential source of radiotoxicity, a potential mitigation to the consequences of accidental scenarios (*e.g.* human intrusion) in the repository evolution with time and reduction of heat load in the repository. However, despite this common generic interest for P&T, different objectives are pursued that can be gathered into three categories:

i) *Sustainable development of nuclear energy and waste minimisation*. The objective to develop sustainable nuclear energy and minimise nuclear waste can be reached through multi-recycling of the TRU as unloaded from LWR in FR and, successively, from FR, if a transition from a LWR fleet to a FR fleet is foreseen. Two options (TRU homogeneous or heterogeneous recycle) can be envisaged and for both, the objective is a stabilisation of the TRU inventory in the reactors and in the fuel cycle, together with the minimisation of the masses sent to the repository (in practice limited to the losses at reprocessing).

ii) *Reduction of TRU inventory as unloaded from LWR*. This objective is related to the management of spent fuel inventories, as a legacy of previous operation of nuclear power plants.

As for reprocessing, a grouped TRU recovery without separation of Pu from MA can be envisaged. To maximise consumption, a U-free fuel (inert matrix) in a fast neutron spectrum device (essentially an ADS) can be envisaged.

iii) *Reduction of MA inventory*. This objective is compatible both with a use of Pu as a resource in LWR for a limited period of time, in the hypothesis of a delayed deployment of fast reactors, and with a sustainable development of nuclear energy, based on the deployment of fast reactors. As far as the transmutation strategy, two options can be envisaged: 1) MA targets, preferably with an inert matrix, to be loaded in critical Pu-fuelled fast reactors; 2) MA fuels in ADS. Pu from LWR will be recycled in MOX-LWR, as foreseen in the double-strata strategy.

Despite the different objectives, the options for P&T implementation show a significant common trunk which allows to draw a consensual European roadmap and also to propose a regional approach of shared fuel cycle installations.

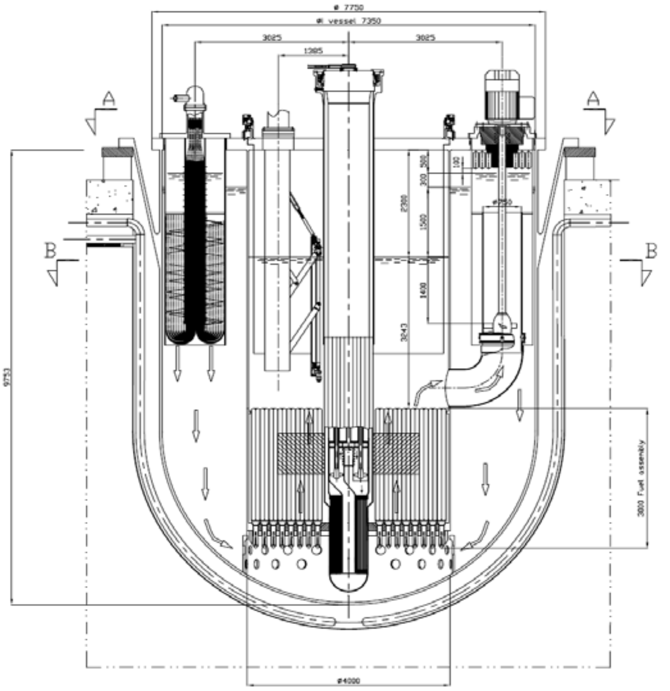
4 Subcritical accelerator-driven transmutation systems

As described in Section 2, the best transmutation performance can be reached with nuclear systems having a fast neutron spectrum. These systems can be critical or subcritical fast reactors cooled with different types of coolants.

As far as Europe is concerned, over the past 15 years the European Commission has supported several projects to develop subcritical ADS to transmute LWR spent nuclear fuels. These projects have addressed the main R&D issues needed for the development of ADS, which comprises the development of an ADS design (including the needed accelerator), improvement of the related nuclear data, the feasibility study of subcritical cores coupled with a neutron source and an accelerator, the dedicated fuel development and the related structural materials and coolant technology development. In particular, after the design activities of the "Preliminary Design Study of an eXperimental ADS" project (PDS-XADS) [6] supported by the FP5 EC programme, the EUROTRANS project [7] is presently the European reference project for development of ADS systems.

The objective of EUROTRANS is the design and the feasibility assessment of an industrial ADS prototype dedicated to transmutation. The work programme includes the advanced design of an approximately 50-100 MW_{th} experimental facility with the aim to demonstrate the technical feasibility of transmutation in an accelerator-driven system (XT-ADS), and a generic conceptual design with a power of several 100 MW_{th} of a European Facility for Industrial Transmutation (EFIT), see Figure 1.

Figure 1: Conceptual design of EFIT



Both facilities foresee the use of heavy liquid metal (HLM) as coolant and as neutron spallation material [8]. As reported in Ref. [9], the thermal-physical and neutronic properties of the HLM lead-bismuth eutectic (LBE) and Pb makes these materials very attractive to be used as coolant in nuclear reactors and in neutron spallation target systems. On the other hand the structural materials' compatibility in terms of corrosion and mechanical resistance poses a major concern for these systems. Therefore, dedicated R&D programmes have been promoted to select and characterise suitable structural materials.

5 Materials challenges and key components of subcritical ADS

The main components of the ADS are the accelerator, the neutron spallation target and the subcritical blanket. The neutron spallation target is located inside the subcritical fast neutron reactor core and provides the external source of neutrons, as needed for a stable operation of the ADS system. From the technological point of view the neutron spallation target represents the most innovative component. The reference neutron spallation solution for the EFIT and the XT-ADS design is the windowless option where the proton beam impinges directly on the LBE. A further option is the window target design, where the proton beam and the spallation material are physically separated by a window usually made of steel. This option has been developed and tested in the frame of the MEGAPIE initiative [10], as is described in Section 5.3.

In parallel to programmes dedicated to design activities, several other research projects have been funded to select and assess the structural materials and develop the technologies associated to ADS. These technologies were mainly related to HLM coolants as Pb and LBE. Chronologically, the first projects were dedicated to screening tests in order to study basic structural material phenomena related to the ADS chemical and nuclear environments. A first structural materials selection was then made for the MEGAPIE target, where the ferritic/martensitic (F/M) steel T91 and the austenitic steel AISI316L were chosen. Finally, both steels are considered as the reference for core components and primary system of the ADS. The main materials properties studied within these projects were their chemical and mechanical resistance to corrosion and their mechanical behaviour under neutron and neutron/proton irradiation. In the sections that follow, the main achievements of the four most relevant projects related to HLM technology and supported by the European Commission in the

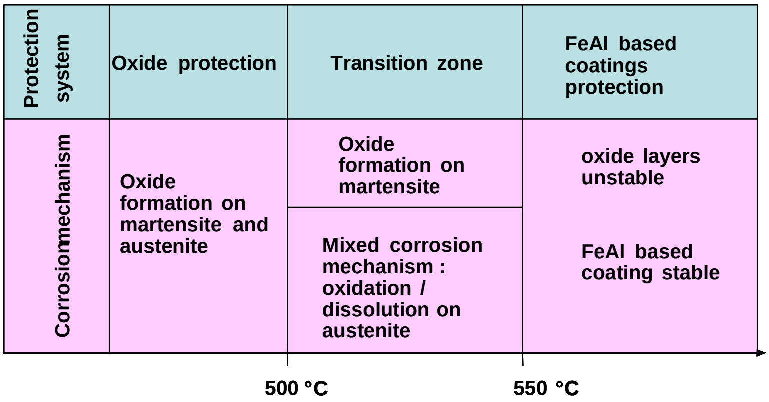
5th (1998-2002) and 6th (2002-2006) Framework Programmes will be summarised. These projects include: *i)* “Technologies, Materials and Thermal-hydraulics for Lead Alloys” (TECLA); *ii)* “Spallation and Irradiation Effects” (SPIRE); *iii)* “Megawatt Pilot Experiment” (MEGAPIE-TEST); *iv)* the domain “Development and Assessment of Structural Materials and Heavy Liquid Metal Technologies for Transmutation System” (DEMETRA) within the EUROTRANS project. While the first three projects have been completed, the DEMETRA domain of EUROTRANS will not be completed until March 2010.

5.1 TECLA

Within TECLA screening tests on several types of austentic and martensitic steels have been performed to study the compatibility of those steels with Pb and LBE and to possibly assess the corrosion mechanism [11]. Almost all TECLA results are obtained from tests performed in stagnant conditions. The main results, summarised in Figure 2, are:

- The compatibility of the materials with the HLM is strongly dependent on the chemical quality of the HLM, where the oxygen activity and the temperature play a major role.
- With an appropriate oxygen activity in the liquid metal a protective (against corrosive attack) oxide scale is formed on steel (T91, EM10, EUROFER, A316L) surfaces; for temperatures higher than 500-550°C, however, the oxide layer becomes non-protective.
- Aluminised coatings tested under similar conditions are effective in avoiding HLM corrosion attack on austenitic and martensitic steels up to 550°C.

Figure 2: Identified corrosion mechanism and corrosion protection systems within TECLA



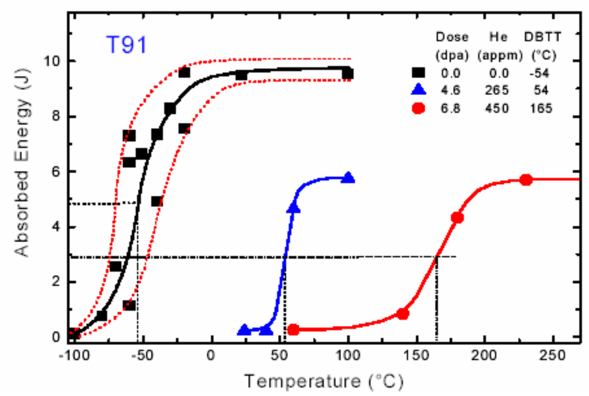
Protection system	Oxide protection	Transition zone	FeAl based coatings protection
Corrosion mechanism	Oxide formation on martensite and austenite	Oxide formation on martensite Mixed corrosion mechanism : oxidation / dissolution on austenite	oxide layers unstable FeAl based coating stable

5.2 SPIRE

The SPIRE project mainly addressed the mechanical behaviour under neutron and neutron/proton irradiation of F/M steels pre-selected as structural materials for the spallation target window or for the structure of the core [12]. The main results obtained within this project are:

- Irradiation under proton/neutron mixed spectrum (SINQ) leads to an increase of hardening with decreasing the irradiation temperature (T < 300°C) with an acceptable ductility for the investigated conditions [dose ~ 12 dpa, He < 1 000 atom-part per million (appm)] (Figure 3).
- To optimise the lifetime of the spallation target, the in-service temperature range would be: 350°C < T < 500-550°C.
- Based on the reactor irradiation results (neutron irradiation), at T < 350°C, hardening and embrittlement induced by the irradiation could reach a prohibitive level. At T > 500°C, besides irradiation effects, corrosion, thermal creep and creep-fatigue contribute to define the upper limit of in-service temperatures.

Figure 3: Ductile to brittle transition temperature of T91 steel irradiated in a neutron/proton mixed spectrum [13]



5.3 MEGAPIE-TEST

The objective of the MEGAPIE-TEST project was to design, build and operate a high power (of the order of 1 MW) neutron source for accelerator-driven subcritical systems (ADS) [14].

In a neutron spallation target, the neutrons are produced via spallation reaction of a high power proton beam colliding with the material of the target. The power of the beam and the specific target material, define important characteristics of the ADS. For instance a 1-2 MW_{th} spallation heavy liquid metal (HLM) target would be required to provide the external source for a subcritical system having a k_{eff} between 0.95 and 0.98 and a core power between 50-100 MW_{th} [15].

In fact, in order to get strong neutron sources, the use of HLM as spallation materials has been envisaged. However, an HLM target to be irradiated in a high-intensity proton beam has never been built. Moreover, it has been generally recognised that the spallation target is an innovative and most challenging component of an ADS, itself an innovative concept which would require a detailed experiment validation. To this end, an international initiative (the MEGAPIE, MEGAwatt PIlot Experiment) was launched in 1999 to build and operate a representative (in terms of power and target material) spallation target, to be realised in the SINQ facility at PSI [10].

The structural materials selected for MEGAPIE were the F/M steel T91 for the proton beam window and the lower liquid metal container and the austenitic AISI 316L steel for the pumps, heat exchanger, main/bypass flow guide tubes, central rod, fill and drain tubes. A scheme of the target is shown in Figure 4.

The MEGAPIE-TEST project has taken advantage of the TECLA and SPIRE projects to assess the selected materials in terms of corrosion and irradiation performances. In addition, specific experiments under MEGAPIE-relevant conditions have been performed to evaluate the target lifetime [16]. On the basis of all experimental results produced it was concluded that the impact of corrosion, irradiation damage and liquid metal embrittelment on the AISI316L steel was relatively low. On the contrary the impact of irradiation damage on the T91 steel was driven by the DBTT shift (see Figure 5) that has been considered as a limiting factor. Indeed, it was concluded that the DBTT shift of the steel should remain below the minimum temperature, 230°C, which corresponds to a maximum of 3.4 Ah (*i.e.* 8-9 dpa). Finally, MEGAPIE has been operated for four months up to an accumulated charge of 2.8 Ah and with availability of the order of 95%. In the future MEGAPIE will be dismantled and an extensive PIE programme will start.

5.4 DEMETRA

Within the EUROTRANS project the austenitic stainless steel AISI316L (vessel and in-vessel components) and the ferritic/martensitic (F/M) steel T91 (core components) have been confirmed as reference structural materials [17], since they have been considered, among the commercial available steels, as the most promising. In order to assess their performance in relevant conditions of both the XT-ADS

Figure 4: Schematic of MEGAPIE target assembly

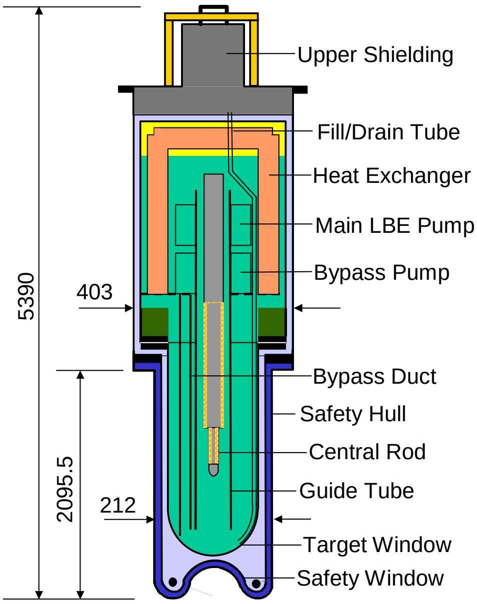
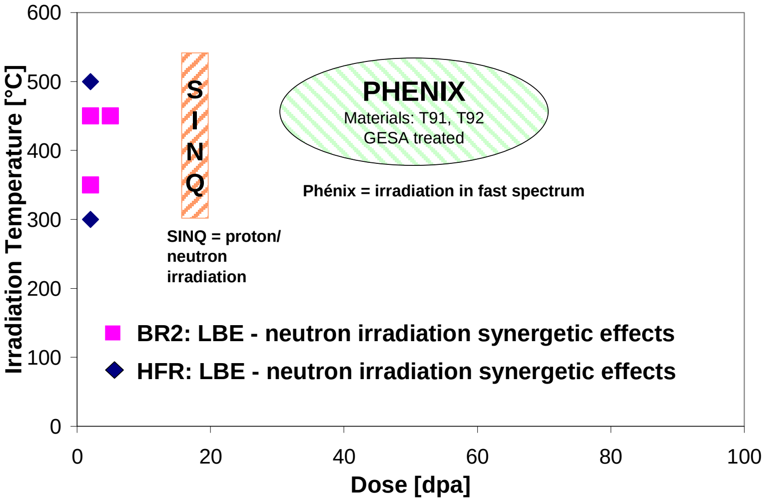


Figure 5: Summary of the irradiation campaign started in the DEMETRA domain



and the EFIT designs an extensive test matrix has been set-up [17]. The test matrix comprises corrosion tests in flowing liquid metal, mechanical tests in HLM and the preparation of irradiation tests in a fast spectrum and in a neutron/proton field in a higher temperature and dpa range with respect to those investigated in the SPIRE project. Moreover, to investigate eventual synergetic effects between the HLM and the neutron irradiation field, dedicated irradiation experiments have been prepared as well, and are currently ongoing.

The main outcome of the DEMETRA domain is summarised in the following sections.

Compatibility with HLM

The experiments performed in non-isothermal HLM loops confirmed that the oxidation of the steels is dependent on the oxygen content in the HLM. Indeed, at relatively low oxygen concentration the corrosion mechanism changes from oxidation to dissolution of the steel elements. Further, the oxygen rate's dependency (expressed in terms of oxide thickness) on the HLM flow velocity, the temperature and the stress level on the steel has been observed. A relation between these parameters to evaluate corrosion rate with physical models is under consideration.

The occurrence of an oxide scale on the structural materials for, *e.g.* the cladding and the heat exchanger might be beneficial to protect the steel against dissolution attack and eventual degradation of the mechanical properties of the steels assisted by the liquid metal [18]. However, a too-thick oxide scale can change the thermal conductivity of the materials, with consequences on the heat transfer of the system and on the thermo-mechanical resistance of the steel [19]. Therefore the evaluation and the understanding of the parameters affecting the oxidation rate could allow defining on optimal range of parameters to keep the oxide scale thickness at an acceptable level.

In parallel a corrosion protection method is developed in order to protect the claddings (made of T91 steel) from corrosion/oxidation. This method consists in spraying Fe, Al based powders on the steel surface and to treat the obtained coating with pulsed electron beam at the GESA (Gepulste Elektronen Strahl Anlage) facility [20]. First results have shown that the Al content in the coating needs to be controlled in order to assure the corrosion protection capability of the coating. Moreover, corrosion tests performed on GESA treated samples in flowing HLM up to 600°C have confirmed the effectiveness of this method [20].

A set of mechanical tests on T91 and AISI316L steels in HLM to investigate liquid metal embrittlement and environmental assisted degradation of the mechanical properties has also been performed. These tests included LCF, tensile, impact and creep to rupture tests. First results obtained show that for high stress level, surface cracks on the steel surface and low protection against liquid metal attack might have a degradation effect on the F/M steel.

Irradiation of structural materials

Figure 5 summarises the parameters of the irradiation experiments ongoing within DEMETRA. The aim of this wide test matrix is to study the mechanical behaviour of the reference structural materials and the behaviour of the corrosion protection barriers exploring a large range of temperature and doses. The BR2 (at SCK•CEN) and HFR (at NRG) experiments shown in this figure are dedicated to the study of the neutron irradiation/LBE combined effect. The irradiation in the Phénix reactor (fast spectrum), the BR2 and HFR reactors has started. Concerning the PIE of these tests, most of them are expected by the end of 2009.

As far as SINQ (PSI) irradiation is concerned, this is to investigate the combined effect of proton/neutron irradiation where a relevant amount of He in the structural material is generated, which corresponds to the typical conditions of a neutron spallation target.

PIE results obtained on the irradiated samples in SINQ have shown that the AISI316L irradiated between 100 and 400°C up to 17 dpa and 1 600 appm of He retains under tensile load a certain ductility, while the T91 steel irradiated under similar conditions (irradiation temperature up to 300°C) shows a certain degree of hardening and embrittlement [21].

6 Materials issues for critical transmutation systems

As already described in Sections 2 and 3, spent nuclear fuel, properly handled, can be transmuted in critical fast reactors. This option has been considered in different projects (CAPRA/CADRA, advanced fuel cycle initiatives and more recently as well in the Generation IV initiative).

As far as the European framework is concerned, a number of European nuclear stakeholders recently issued a vision report to establish the Sustainable Nuclear Energy Technology Platform (SNE-TP) [22]. The SNE-TP is preparing the European Strategic Research Agenda (SRA) which includes the objective of preparing the development of sustainable nuclear systems for the future, including the advanced fuel cycle. To reach this objective fast neutron reactor systems are under consideration.

In this context, the key material issues identified for the development of fast neutron transmutation systems are those related to core components, which are subjected to the most severe in-service conditions. The structures of the fuel subassemblies, such as fuel cladding and wrapper tubes, must bear during operation high levels of radiation damage, high temperature and coolant effects (Na, Pb or Pb-Bi eutectic, He). At present, high-Cr F/M steels are considered as the reference structural materials for the components of the primary systems (as proposed for the Japanese SFR [23]), for the cladding (as proposed for the heavy liquid metal-cooled systems [24,7]) and for the vessel (as proposed for the gas-cooled systems [25]). The preference for F/M steels in all these applications is mainly related to the better thermal properties (higher thermal conductivity and lower thermal expansion), better dimensional stability under irradiation [26] and higher compatibility to heavy liquid metals [17], when compared to austenitic steels.

However, the mechanical strength of the considered high-Cr F/M steels is limited to temperatures as high as 550°C [26] and, on the other hand, the use of austenitic steels (as *e.g.* cladding material) would limit the desired high burn-up range, due to their low resistance to radiation-induced swelling [26]. These are the main reasons which have motivated the materials science community to study and develop new structural materials which could cope with the objective of enhanced efficiency of the innovative reactor concepts.

In Europe, the European Commission has launched in its 7th (2007-2013) Euratom Framework Programme a research project to study and qualify commercially available materials and for the longer-term prospective to develop and qualify new materials and corrosion protection barrier. This project has been named “Generation IV and Transmutation Materials” (GETMAT) [27]. For the high temperature strength material, within the GETMAT project, priority has been given to the development and study of Fe-Cr ODS alloys, since these show favourable properties for high temperature and high burn-up applications. Moreover, ODS alloys would allow to increase the operating temperature above 550°C and in contrast to austenitic steels and Ni-based alloys, the ODS steels do not show high temperature He-embrittlement or irradiation-induced swelling [28]. Results of irradiation experiments up to 35 dpa at 330°C indicate good irradiation resistance, with tolerable irradiation hardening and little loss of ductility [29].

In summary, the focus of the GETMAT project has been placed on the development and characterisation of ODS and high Cr F/M steels, their welding/joining and their qualification, in terms of mechanical and corrosion resistance under appropriate conditions. An extensive post-irradiation examination (PIE) programme, taking advantage of ongoing irradiation experiments (see Section 5) will contribute to the assessment of the two classes of materials. Finally, an important part of the GETMAT project is dedicated to the fundamental understanding of the physical mechanism at the basis of the materials behaviour under irradiation through multi-scale modelling activities and experimental validation.

The five-year GETMAT project began in February 2008 and the final goal of this project is to contribute to the selection and qualification of structural materials for the innovative reactor systems considered in the framework of Generation IV and advance fuel cycle initiatives.

7 Conclusions and perspectives

There is clear evidence from numerous past studies that the best transmutation performance can be reached with fast neutron critical (cooled with Na, Pb or He) or subcritical reactor systems (cooled with Pb or LBE). Moreover, the transmutation performance of the fast neutron systems is not significantly affected by the coolant type. The objectives of management with P&T of the spent nuclear fuel as unloaded from the LWR, can be classified into three main categories, *i.e.* sustainable nuclear development with waste minimisation, reduction of transuranics inventory and reduction of minor actinide inventory. Depending on the envisaged objective the preferred system can be either a critical or a subcritical reactor system.

In Europe both reactor types are being developed, as the European Commission has supported design-oriented programmes to develop ADS subcritical transmutation systems cooled with Pb and LBE. In parallel, several projects to assess the structural materials and to develop key components such as the neutron spallaton target have been created. Most of these projects have been successfully completed or are close to completion and have assessed the most promising commercially available steels for the ADS. The experimental results made evident the limits of the mechanical and corrosion resistance of these steels in the considered temperature and irradiation range. Moreover, the design, fabrication and operation of key components such as the neutron spallation target have been demonstrated.

The future implementation of transmutation of the spent nuclear fuel is driven in Europe through the Sustainable Nuclear Energy Technology Platform, where a Strategic Research Agenda is under preparation. In the SRA it has been recognised that whatever the reactor system selected to transmute the spent fuel, the materials research is a key area. Indeed, for the envisaged systems, advanced materials and their assessment has been considered as an essential issue. The European Commission has therefore launched a cross-cutting research project on structural materials for Generation IV and transmutation systems. This project, recently launched, has as its objective to complete the assessment of F/M steels and to develop ODS steels which have improved high temperature strength and therefore could be utilised as core components for the more advanced and innovative transmutation systems, where higher temperature and higher burn-up have been envisaged.

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Material development in lead-bismuth spallation target system

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Abstract

In the conceptual design of lead-bismuth spallation target system a depositing power density is hundreds W/cc and then a plate thickness is limited to 2-3 mm in the proton beam window as design results considering the balance of thermal and mechanical loadings. R&D to know materials performance and its limit for usage applying to the beam window are conducted in JAEA at proton irradiation material test and lead-bismuth flowing loop test by using JLBL-1, 2 and 3. Spallation target material irradiation experiment showed properties due to high-energy proton bombardment. Ductility reduction and off-set stress increase were occurred in austenitic steels. Fracture mode change in ferritic-martensitic steels, such as 8-9 Cr steels, was obtained by other institutes and suggested as points to be considered in material design work. Fatigue test is under way in the hot cell. Microstructure analyses found that many bubbles exist in the materials. The relation between mechanical strength and nano-scale observation results is studied now. Corrosion of the materials under the lead-bismuth flows showed local erosion corrosion. This was not fully explained in our loop so that a visualisation technique has been developed by using ultrasonic Doppler method and surface vitalising techniques to inspect a local flow condition. A thermal fluid experiment was conducted to determine the heat transfer performance at the beam window model, which will affect the thermal loading magnitude. An experimental formula was established for design work.

Introduction

In the conceptual model of nuclear transmutation device in which accelerator plays a role to produce an external neutron source by making high-energy proton beam, one of the issues is to design a beam window with a reliable lifetime and taking into consideration the fuel exchange plan. The beam window is put on high-energy proton beam bombardment under the high temperature lead-bismuth eutectic (LBE) flow. From the point of view of material properties it is well known that not only irradiation but also liquid heavy metal will make material brittle.

In conceptual design work LBE spallation target system a depositing power density is hundreds W/cc and a plate thickness is limited to 2-3 mm at the proton beam window as a result of balance in thermal and mechanical loadings [1,2]. R&D has been ongoing in the field of proton-irradiated materials test using controlled facilities and LBE flow control test by using JLBL-1, 2 and 3: there are LBE flow devices for materials corrosion, fluid in the target model and heat transfer at the beam window model, respectively.

Spallation target material irradiation experiment showed influence due to high-energy proton bombardment on how ductility reduces and how off-set stress increases with increasing displacement per atom (dpa) in austenitic steels. For the case of usage in ferritic-martensitic steels, such as 8-9 Cr steels, ductility brittle transition temperature (DBTT) shift is a design matter under the irradiating environment. Other organisations conducted small punch tests and Sharpy impact test after proton irradiations and showed how DBTT increases with increasing dpa.

Fatigue test is under way in the controlled facilities. Microstructure analyses showed that many bubbles exist in the austenitic and ferritic-martensitic steels homogeneously in the matrix. The relation between mechanical strength and microstructure observation has been inspected. Corrosion of the materials under the LBE flows showed local erosion so that a visualisation method was developed using an ultrasonic Doppler method and surface vitalising techniques. Normally measurement is conducted as low as possible because of transducer problem but wetting between LBE and austenitic steel is wrong at low temperature. A thermal fluid experiment was conducted to determine the heat transfer performance at the beam window, which will affect the thermal loading magnitude. Experimental formula was established for design work.

Proton irradiation experiment

Spallation target irradiation experiment started in JAEA in conjunction with development for neutron source project. PSI provides proton irradiation field in the SINQ target [3]. JAEA prepared specimens to the programme organiser. In total, a couple hundred samples from JAEA were irradiated. Tensile and bending fatigue machines were used for post-irradiation experiment. Such machines were newly developed for handling small specimens. Elongation in the gauge volume was measured directly by the optical microscope. This is an essential technique to know the true strain of the specimens. Specimens include austenitic steels and ferritic steels, which will be applied to liquid target vessel as the beam window. On the other hand, tungsten, tantalum and other refractory metals were also included for applying to solid spallation target. Our emphasis has been toward austenitic steel.

Tensile property of austenitic steel

The tensile property of JPCA (14Cr-16Ni-2.3Mo-0.22Ti, Fe-Bal, wt.%) was studied at RT and 250°C, and at 5.7 to 10.1 dpa, respectively [4,5]. Proton/neutron irradiation caused considerable irradiation hardening and degradation of ductility for JPCA-SA. The increase in 0.2% offset yield stress (YS) of irradiated specimens is within the fission neutron data band at lower dose. At higher dose, the increase in YS of JPCA-SA shows a tendency to exceed the upper bound of the fission neutron data band. Because of strain localisation, uniform elongation falls to very low values in spite of large total elongation and large reduction of area. By means of the true strain to plastic instability, it was shown that the irradiated specimens still have ductility. The fracture surface of irradiated tensile specimens at 10.1 dpa, tested at RT and 250°C are ductile, respectively.

Microstructure observation was undertaken for unirradiated specimens, 5.7 dpa/150°C, 7.9 dpa/230°C, 10.1 dpa/285°C and 19.5 dpa/420°C. A high density of small dislocation loops formed after irradiations. The increase of yield stress and loss of elongation on irradiated samples can be explained by the formation of these high-density small dislocation loops. Density changes are small compared to conventional neutron irradiations. Very high density of small He bubble formation at very low temperature and dose compared to conventional neutron irradiations. He bubble behaviour shows quite different manner compared to neutron irradiation with low He production rate. The density of He bubble does not increase so much and the size also does not change with irradiation dose and temperature. At highest dose 20 dpa and at 400°C He bubble agglomerated near twin boundaries, which was not observed for lower dose and temperature. Mechanical tensile test and fracture surface observation are scheduled for highest dose specimen up to 20 dpa.

Tensile property of ferritic-martensitic (FM) steel

The advantage in using FM steel is its high strength, and the disadvantage that it is less ductile than austenitic steel for specimens irradiated below 350°C at SINQ. There is a discussion about ductility recovery process by proper heat treatment. Principally it is possible and needs considerations on the effect to other devices and how to do. Welded specimens of F82H (8Cr-2W) TIG and EB showed little ductility in our test. This suggests that it is better to keep away welded part from proton exposure. There is a material issue on ductility brittle transition temperature (DBTT) shift due to irradiation. If DBTT is over the temperature at normal operational condition of lead-bismuth (LBE), a beam window material may have a risk to fracture in a brittle way. This is undesirable event for getting reliability. DBTT shift is proportional to helium production rate up to 800 appm [6]. Then He production rate will be critical for usage of FM steel.

Jean Henry showed tensile property of 9Cr-1Mo steels, EM10 (9Cr-1Mo) and T91 (9Cr-1MoVNb). Materials were irradiated at SINQ up to 20 dpa [7]. Below 16 dpa tensile fracture surface was ductile but over this value it was brittle and some specimens broke at elastic regime. Corresponding He production rate to 16 dpa is 1300-1500 appm. Fracture surface was occupied by inter-granular grain more than 50%.

Jia Xuejun observed microstructure of F82H irradiated up to 20 dpa [8]. Bubbles were observed in the matrix as well as M23C6 precipitated carbide. Large bubble around the size of tens nm adjoined precipitated materials.

Fatigue property

Fatigue data of specimens irradiated by proton is limited. Fatigue specimens were irradiated at SINQ and some of them have already been tested. Specimen size is very small as 16 mm in length, 2 mm in width and 0.4 mm in thickness. Specimens are loaded by deflection-controlled mode. There is no difference between unirradiated and irradiated specimens. This is explained by the fact that the cycles to crack initiation were not affected by irradiation in very thin plate. A bending fatigue test for different materials is ongoing. The stress level applied to the specimen surface is 100-200 MPa.

MEGAPIE sample

After a successful four-month operation and the following cooling period, the MEGAPIE target is being readied to provide samples to various laboratories. Unused T91 specimens were dispatched to the partners for assuring the one of machine performance: micro-hardness test was done to provide data for comparison. Samples will be transported next year and PIE will be done over the next two years. The results will show materials quality after operation less than 330°C LBE temperature.

Lead-bismuth flow control

In JAEA there are three major LBE flow loops: those are JLBL-1, 2 and 3 dedicated to do experiment on corrosion, flow measurement and heat transfer of the beam model, respectively.

Corrosion test at JLBL-1

We have tested totally five runs and total running time achieved more than 18 000 hrs. The loop consists of SS316. LBE inventory is 22 L, maximum operation temperature is 45°C and LBE flow rate is

5 L/minute, which equal to 1 m/s at high temperature tube specimen with 10 mm in diameter and 0.2 m/s at lower temperature specimen with 24 mm in diameter. Flow rate is measured by EMF and LBE is driven by EMP.

First period

Test condition was 400 to 450°C during 3 000h rs. Remarkable corrosion erosion was found at the specimens exposed to 450°C [9]. Corrosion depth achieved was about 0.1 mm. It was surprising to know too much higher corrosion rate. And also flow meter indication was decreased with increase of time by about 50%. We inspected EMP and EMF. It was seemed that one of devices might be wrong. Results showed that the pump was mainly responsible for flow rate reduction. Channel was squeezed by precipitation of Fe-Cr crystal. EMP utilised magnetic field. Induced current and induced magnetic field makes driving force to liquid LBE in the channel. Fe-Cr magnetic crystals were trapped in the magnetic field effectively. Solution is to widen channel gap with considering power reduction for LBE flow. We also inspected flow meter. LBE flow in the magnetic field produces output potential. The potential was measured at the difference between two electrodes, which gave the maximum value. Then it was found that the electrodes were flattened by LBE flow. Further inspection revealed that surface oxide with having high electric resistance might disturb induced current flux and potential. So the additional R&D was started in JLBL-2 in order to validate the relationships between input current to the coil and LBE flow rate. The details are reported elsewhere.

Second period

Inside specimens were polished because an inspection of SS316 tube revealed that grain boundaries of inner surface were creviced by the cold working for making tube, heat treatment in air and the following acid washing to remove oxide layers. Polishing could not remove damaged layers completely but corrosion rate reduced to 20-30 micron m at the same operating condition with the first run. For this period corrosion loop was equipped with filtering devices that consisted of metal stripe and magnet. Inspection of filter stripe showed that a number of new Fe-Cr crystals were trapped there. It works at removing precipitations or at accelerating dissolution.

Third period

In ADS design study the temperature difference of LBE is over 50°C but under 100°C. Operating conditions were set to the same as for the second run, but the maximum temperature difference was 350 to 450°C. The results showed again erosion corrosion for specimens in the high temperature part. Cross-section of tube indicated a reduction of thickness and corrosion along grain boundaries. At the joint part where cross-section areas are both increased and decreased surfaces are eroded. Flow simulation suggests that secondary flow is apparently responsible for the erosion. The Reynolds number at the specimen is 50 000. X-ray analyses were done again for the solidified LBE remaining on the specimen. We never recognised precipitated Ni as crystal. But diffused Ni into SS316 and dissolved Ni into LBE have been analysed by EDX, EPMA and ICP. We thought that solubility of nickel is very high comparative with Fe and Cr, estimated to be a couple of per cent. However, needle-type thin precipitation was found on the surface of solidified LBE. By using special techniques a small portion of LBE was successfully cut by FIB and analysed by FE-SEM. It is found to be Ni-Bi precipitation, and seems to be an inter-metallic compound. The Ni-Bi precipitations were found not only at the high temperature part but also at the low temperature part. The reason of severe reduction in thickness seems to be the temperature difference. It accelerated to make Fe-Cr crystal from dissolved LBE. And crystal may have a function of polishing particles. It was decided to change all LBE in the loop. Almost all the LBE was pumped out from the loop and replaced by new LBE solder bars. Samples were collected from the used one. ICP analyses were done to inspect solubility of Fe, Cr and Ni. Fe and Cr were saturated in the used one but Ni did not achieve to the value IPPE reference. It is concluded that Ni precipitated in the cooling process, depending on temperature difference.

Fourth period

Our programme stepped up to the alternative materials: F82H. It is developed by fusion community and characterised by low activation ferritic steel. During this period not only corrosion test but also oxygen sensor test were involved. So that temperature condition was varied in sequential programme; first in isothermal conditions at 300, 350, 400 and 450°C and then set up to a normal condition described by the same with third period: 350-450°C. Ferritic steel was very reliable. Oxygen concentration was 10^{-9} to 10^{-6} wt.%. SEM observation showed different manner with SS316. There is no oxide layer on the surface by EDX analyses. At the expansion part of tube somewhat reduction of thickness was observed. In comparison with other F82H data, 10 to 20 micron m oxide layer had been expected under the possible oxygen concentration.

Fifth period

F82H + GBE specimens were put in the loop. Temperature conditions were set to 350-450°C. GBE specimens were set to the centre position of the tube cross-section. The shape was 10 mm square with 1 mm thickness. GBE specimens were provided by Tohoku University. The specimen was made from SS316L. GBE and the parent material were tested. GBE specimens showed remarkable corrosion erosion resistance in comparison with parent material. Oxygen concentration during the period varied from 10^{-8} to 10^{-4} wt.%. There was no active oxygen control but loop plenum was covered by Ar gas with 99.95% purity.

Flow profile measurement at JLBL-2

JLBL-2 is a target flow model. The shape is cylindrical type with 7 cm in diameter and 1.5 m in length. In the cylinder there are two channels, which are annular type and duct types arranged along the centre line. LBE flows counter direction at the outer and inner channels. R&D was done for measuring flow velocity profile by ultrasonic Doppler method. Emphasis was put on how to get good transmission of sonic wave between austenitic steel and LBE [10]. In the LBE sonic energy is so easy to disperse by the reflection that possible measuring depth is within 10 cm for the case of 4 MHz input transducer. In principle the Doppler method measures the velocity of the particle in the fluid. Particles are a different substance from LBE but have nearly the same density, sometime impurity including in the LBE like slugs and oxide particle, or gas bubbles produced by the flow. We could measure the velocity profile of LBE in the target model without help of additional particle. This method will be able to apply to the study of local flow distribution. At the same time we investigated EMF because in JLBL-1 operation as mentioned before output was reduced by some reason. So that analyses of electric potential was done in order to know the effect of oxide layer adhered on the surface of the flow channel. As oxide layer has a high electric resistance, it will affect potential distribution and intensity. The results are applied to the advanced system of EMF at JLBL-2.

Heat transfer measurement at JLBL-3

There are analytical results on Nu number on the surface of round body in the flow but nothing about the shape of the beam window of the conceptual model in ADS, which is hemispherical shape connected with cylindrical tube. The shape of hemisphere itself is an issue to design reliable stress distribution against hydrostatic pressure of LBE. The beam window is submerged at 6 m in depth LBE. The pressure is 6 MPa there.

In order to measure the Nu number at the beam window, JLBL-3 was constructed. It is capable of flowing 500 L/minute. The beam window model is 5 mm in thickness and 90 mm in diameter. It is equipped with a 6 kW heater in order to heat one sidewall surface uniformly. The other side is cooled by LBE flow. A temperature difference was measured by TCs, which were varied near the surface of the both sides. LBE temperature is maximum 450°C and the Re number is maximum 500 000 for the beam model diameter. LBE was driven by mechanical pump. Temperature fluctuation was observed at the beam window model. Details are being studied. Local Nu number along the beam window model was formulated [11].

Summary

In proton irradiation materials experiment we emphasised austenitic steel. It is found that it has ductility up to 10.1 dpa and further investigation will be done up to 20 dpa. Microstructure observation showed that small bubbles are produced homogeneously in the materials. Further investigation will be done to know a relationship between mechanical property change and bubbles existence. Fatigue test data is very limited so that we will continue to test samples.

In LBE control study we have tested austenitic stainless steel and ferritic steel: SS316 and F82H at JLBL-1. Active oxygen control was not done but high pure Ar gas covered the loop plenum in order to avoid LBE from air ingress during operation. Oxygen concentration was varied in the range of 10^{-9} to 10^{-4} wt.%. Flow rate of LBE is 1 m/s in average. Temperature controlled to 450°C maximum with a temperature difference of 50 to 100°C. F82H was very stable: oxide layer was not observed and to grain drop was not also observed. SS316 tube with grain boundaries damage due to inadequate heat treatment prior to the test showed corrosion along grain boundaries and grain drop. Conclusively the surface that will contact with LBE must be polished before usage. As there is no corrosion at 400°C and good quality material will endure to usage of austenitic steel at 450°C [12].

In LBE flow measurement ultrasonic Doppler method is effective to visualise. An application, however, to higher temperature is desirable through the enhancement of transducer performance. Wetting of austenitic steel with LBE is not effective at lower temperatures, but vitalising techniques make measurement possible.

In heat transfer experiment we found a fluctuation of temperature at the beam window model. We are investigating these phenomena, which have a close connection with LBE fluid condition.

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Evaluation of the corrosion resistance of FeCrAl alloy in stagnant LBE with active oxygen control

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Abstract

Lead and lead-bismuth eutectic (LBE) alloy have been increasingly receiving attention as heavy liquid metal coolants (HLMC) for future nuclear energy systems. The compatibility of structural materials and components with lead-bismuth eutectic liquid at high temperature is one of the key issues for the commercialisation of lead fast reactors. In the present study, the corrosion behaviors of commercial iron-based alumina-forming alloy (Kanthal®-AF) and austenitic steel (SS316L) as well as 9Cr ferritic/martensitic steel (T91) were investigated through exposure to stagnant LBE environments at 500°C for 500 hours. After exposure, the thickness and chemistry of the oxide layer on the specimens were analysed using scanning electron microscopy (SEM), scanning transmission electron microscopy (STEM) and energy dispersive X-ray spectroscopy (EDS). As a result, the oxide characteristics and the oxidation resistance were compared. Nano-structural analysis results showed that chromium sulphide precipitates present at oxide/substrate interfaces on Kanthal®-AF make the surface oxide layers unprotective. To evaluate sulphur effects on the corrosion behaviour of Kanthal®-AF in LBE, H_2 anneal heat treatment was conducted at 1 200°C for 100 hours with flowing 99.999% H_2 gas. Then, the corrosion behaviours of Kanthal®-AF, with and without a prior H_2 anneal heat treatment, were investigated in stagnant LBE at 650°C. Analysis results show that the H_2 annealing significantly improve the protectiveness of oxide scale.

Introduction

Lead-bismuth eutectic (LBE) alloy is one of the coolant candidates for the fourth-generation (Gen-IV) small- and medium-sized reactor (SMR) nuclear systems because of its low melting temperature, high boiling point, excellent chemical stability, no fire or explosion and neutron transparency. In addition, LBE has high capability for natural circulation [1]. In recent years, lead and LBE have attracted greater attention and increasing R&D efforts world wide, due to global interests firstly in partition and transmutation, and secondly in nuclear systems utilising fast neutron [2,3]. In the same respect, the Nuclear Transmutation Energy Research Center of Korea (NUTRECK) of Seoul National University in the Republic of Korea is developing an innovative technology to burn spent nuclear fuels into low-intermediate level waste by utilising the LBE-cooled PEACER [4] and its small modular reactor design, PASCAR [5].

Those LBE-cooled nuclear energy systems, however, have been exposed to several issues arising from the limitation of system life due to the relatively low corrosion resistance of structural materials in the LBE environments. Since the system lifetime of advanced liquid metal-cooled nuclear power systems including PEACER and PASCAR are designed to be beyond 60 years based on the advanced design technology, the long-term corrosion behaviours have been extensively studied to investigate the corrosion performance of structural materials and improve the materials life performance world wide.

From a previous study by Zhang and Li and as listed below [6], general conclusions of corrosion behaviour of conventional austenite stainless steels and ferritic/martensitic steels (FMS) as candidates of structural materials in LBE environments have been well reviewed and summarised:

- At very low oxygen levels ($<10^{-8}$ wt.%), both austenitic and martensitic steels in LBE are subject to the dissolution of alloying elements even at temperatures as low as 400°C.
- In the temperature range of 300-470°C, protective oxide films can form on the surface of austenitic and martensitic steels at oxygen levels above 10^{-8} wt.%. Thin surface oxide films can protect the steel elements from the rapid dissolution into LBE. Protective layers on martensitic steels are much thicker than those on austenitic steels. Generally, the oxide films on martensitic steels have a duplex structure: an outer porous layer of magnetite and an inner barrier layer of Fe–Cr spinel that grows at both interfaces: the LBE/oxide interface and the oxide/steel interface. In contrast, the oxide films on austenitic steels are monolithic and mostly composed of Fe–Cr spinel at lower temperatures.
- It has been confirmed that austenitic stainless steels suffer from severe corrosion attack in LBE at temperatures above 500°C, while ferritic/martensitic steels suffer from severe oxidation. For temperatures above 550°C, accelerated dissolution of austenitic steels occurs, making these steels unsuitable without special surface treatments. For martensitic steels, very thick oxide layers are formed on the surfaces at temperatures above 550°C. This indicates that long-term applications of martensitic steels may required extensive coolant filtering since thick oxides are not very protective and will become released.

For long-term and high-temperature (>500°C) applications, therefore, Al-containing alloys and Al-coated steels have recently been investigated [7-11]. Coating surfaces of FeCr steels with FeCrAlY after homogenisation by melting with the GESA pulse has shown an excellent corrosion protection at up to 650°C in LBE [7-9]. Impervious thin alumina scales were formed to protect steels from dissolution attack by LBE as well as from internal oxidation.

In the present study, the corrosion behaviours of commercial iron-based alumina-forming alloy (Kanthal®-AF) were investigated to understand the fundamental procedures of corrosion by exposure to stagnant LBE followed by nano-structural examination of surface film and adjacent metal substrate. To compare with conventional FeCr alloys, the corrosion behaviours of austenitic steel (SS316L) and 9Cr ferritic/martensitic steel (T91) were also investigated under the same conditions.

Experimental

Three types of alloy were selected in this study; their chemistry is provided in Table 1. Kanthal-AF® which is manufactured by Sandvik is an iron-based alumina-forming alloy containing small amounts of yttrium and zirconium.

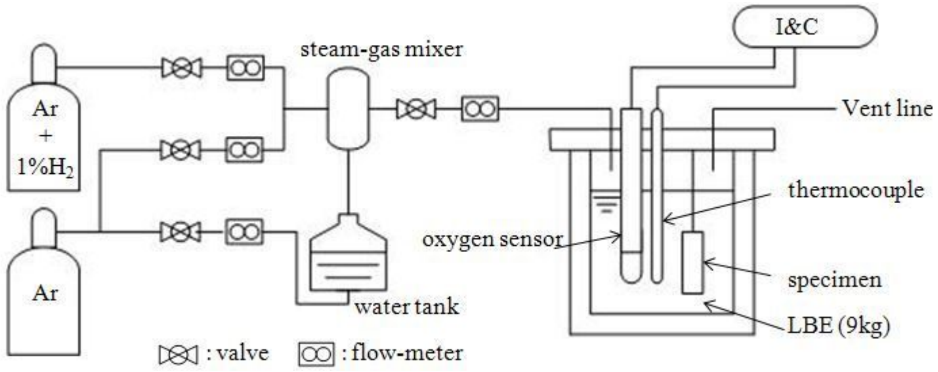
Table 1: Chemical composition of test materials [wt.%]

Materials	Fe	Cr	Al	Ni	Mn	Mo	Si	Ti	Y	Zr	S
T91	Bal.	8.6		0.23	0.43	0.95	0.3				
SS316L	Bal.	17.3		12.1	2.8	2.31	0.3				
Kanthal®-AF	Bal.	22.3	5.1		0.17		0.21	0.07	0.05	0.07	0.004

All specimens were machined to plates with the dimensions of 15 mm in length, 8 mm in width and 1 mm in thickness. Before the exposure in LBE, each specimen was mechanically polished by abrasive SiC papers down to 600 grits with water cooling and then ultrasonically cleaned with acetone and ethanol, respectively, before the final drying.

The corrosion tests were carried out at 500°C for 500 hours in static LBE cells that are connected to an oxygen control system (Figure 1). An alumina crucible was used as the LBE container in each oxygen-controlled cell. The quantity of LBE in the alumina crucible was nominally 9 kg in weight. During the test, oxygen concentration in LBE was controlled over the range of 10^{-6}~10^{-5} wt.% by flowing H_2/H_2O gas with regulated composition and flow rates, and monitored by a yttria-stabilised zirconia (YSZ) oxygen probe that was hermetically sealed using the electromagnetically swaged metal-ceramic joining method for long-term applications [12]. Oxygen-saturated bismuth (Bi/Bi_2O_3) was used as a reference reaction couple that is placed inside YSZ.

Figure 1: Schematics of stagnant corrosion test apparatus equipped with oxygen control system



To evaluate the effects of sulphur impurity in FeCrAl alloy on corrosion behaviors in LBE, H_2 annealing heat treatment was conducted at 1 200°C for 100 hours with flowing 99.999% H_2 gas to remove sulphur from the alloy. Then, the corrosion behaviors of Kanthal®-AF, with and without a prior H_2 annealing heat treatment, were investigated in stagnant LBE at 650°C.

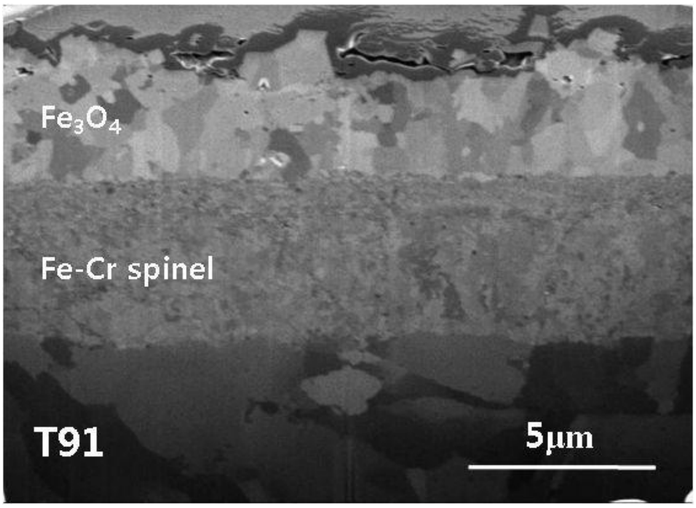
After the experiment, the surface layers of each test specimen were examined using field emission-scanning electron microscopy (FE-SEM) and scanning transmission electron microscopy (STEM) couples with energy dispersive X-ray spectroscopy (EDS). Before the nano-structural analysis, the tested specimens were washed in a hot glycerine bath at about 170°C to remove residual LBE. To analyse the cross-section of surface oxide layers on the specimen, the specimens were ion-milled by focused ion beam (FIB) and TEM specimens were also machined by FIB.

Results and discussion

After exposure to 500°C stagnant LBE for 500 hours, no corrosion damage by dissolution of steel components into the liquid metal (liquid metal corrosion) was detected for all alloys.

A duplex layer oxide film formed on the surface of T91. As shown in Figure 2, the outer oxide layer is Fe_3O_4 while the inner oxide layer is Fe-Cr spinnel. The thickness of oxide layer is about 10 μm. The results of STEM/EDS analysis for the SS316L specimens are as shown in Figure 3(a) display thin (~100 nm) and continuous Cr_2O_3 formed on the surface of the specimen and about 1 μm thick Fe_3O_4 formed on the chromium oxide layer. These results correspond to previous studies as mentioned in the introduction.

Figure 2: Focused ion beam image of the cross-section of T91 after exposure to 500°C stagnant LBE for 500 hours



In the case of Kanthal®-AF, relatively thin (100~200 nm thick) continuous Al_2O_3 layers were observed to form on the surface of the specimen. As shown in Figure 3(b), it was observed that the existence of the chromium sulphide (CrS) island at the Al_2O_3 layer/substrate interface. These Cr and S co-segregations in the interface are often observed for FeCrAl alloys [13-15]. The major effect of S segregation was found to influence interfacial pore formation and to deteriorate the oxide scale adhesion properties. Oxides on the S-segregated region are believed to initiate cracks more easily and allow oxygen ingress to form new oxide with the alloy, thus giving rise to a higher oxidation rate under the gaseous environments.

Figure 4 shows three difference oxide characteristics of Kanthal®-AF after exposure to 500°C stagnant LBE for 500 hours. Thin and protective Al_2O_3 was formed on the surface and the region was not affected by S segregation [Figure 4(a)]. Two different regions were affected by S-segregation. One region consists of outer Al_2O_3 and the chromium sulphide island [Figure 4(b)]. The chromium sulphide transformed to Cr_2O_3 and porous Fe_3O_4 in the other region [Figure 4(c)].

In the LBE environment, Cr and S co-segregation with forming chromium sulphide would affect the oxide growth and dissolution mechanism by LBE as follows.

As the oxygen concentration at the scale region increases by inward diffusion of oxygen through the outer Al_2O_3 layer, the chromium sulphide is possibly getting unstable, because chromium sulphide (CrS, Cr_2S_3) is thermodynamically unstable compared with Cr_2O_3. If LBE penetrates through the defect of Al_2O_3 layer and then contacts with the chromium sulphide, these unstable chromium sulphides can be dissolved by LBE. And the chromium sulphide can also act as an obstacle to the diffusion of Al+ ion that can heal the defect of the outer Al_2O_3 layer. For these reasons, chromium sulphide formed at the oxide/substrate interface due to co-segregation of Cr and S makes the oxide scale less protective.

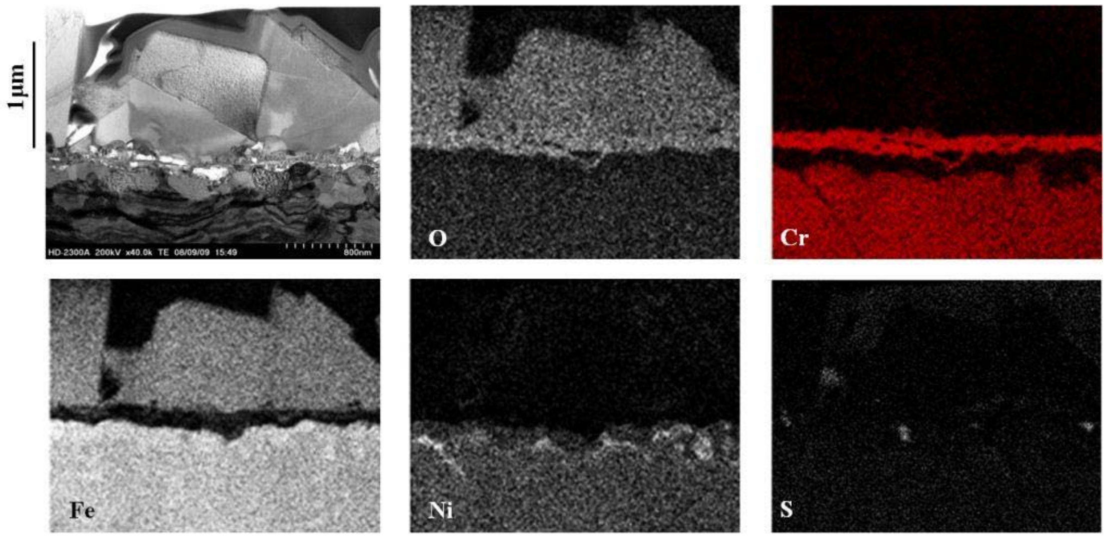
To evaluate the effects of sulphur impurity in FeCrAl alloy on corrosion behaviours in LBE, a H_2 annealing heat treatment was conducted at 1 200°C for 100 hours with flowing 99.999% H_2 gas to remove sulphur from the alloy. Then, the corrosion behaviours of Kanthal®-AF, with and without a prior H_2 annealing heat treatment, were investigated in stagnant LBE at 650°C for 500 hours.

As-received Kanthal®-AF containing 40 ppm sulphur impurity damaged by LBE after exposure to stagnant LBE at 650°C for 500 hours is shown in Figure 5(a) and Figure 6(a). The H_2 anneal heat-treated Kanthal®-AF, however, showed a shiny surface. A thin (100~200 nm), protective oxide layer formed on the heat-treated specimen prevented corrosion damage by dissolution of steel components into LBE as shown in Figure 5(b) and Figure 6(b).

Analysis results show that the H_2 annealing significantly improved the protectiveness of the oxide scale in the FeCrAl alloy.

Figure 3: STEM images and maps of cross-sectional chemical composition obtained by EDS after exposure to 500°C stagnant LBE for 500 hours

(a) SS316L



(b) Kanthal® AF

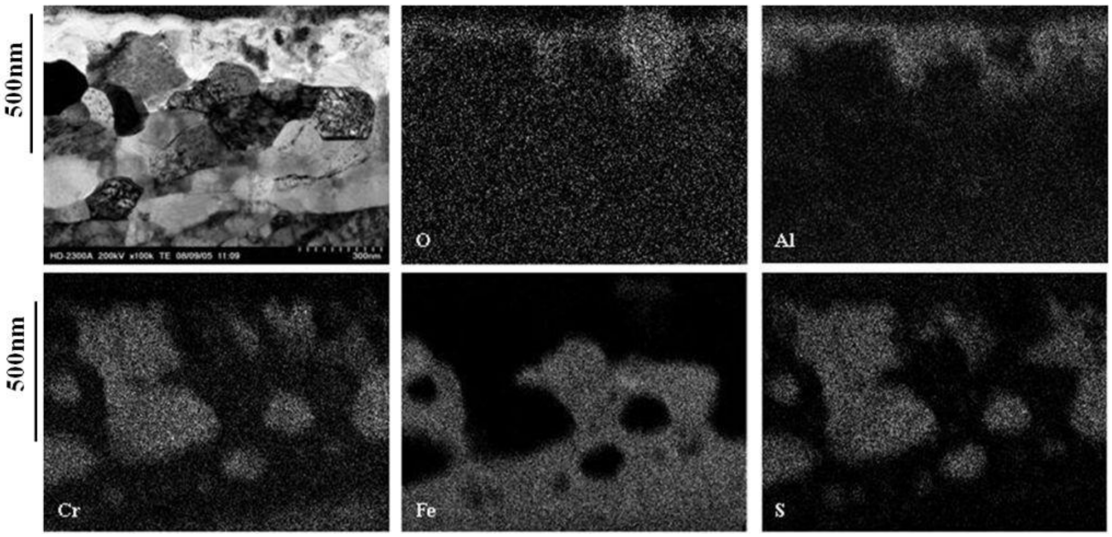


Figure 4: STEM images and maps of cross-sectional chemical composition obtained by EDS for various region of Kanthal®-AF after exposure to 500°C stagnant LBE for 500 hours

Blue: Al, green: Cr, red: Fe

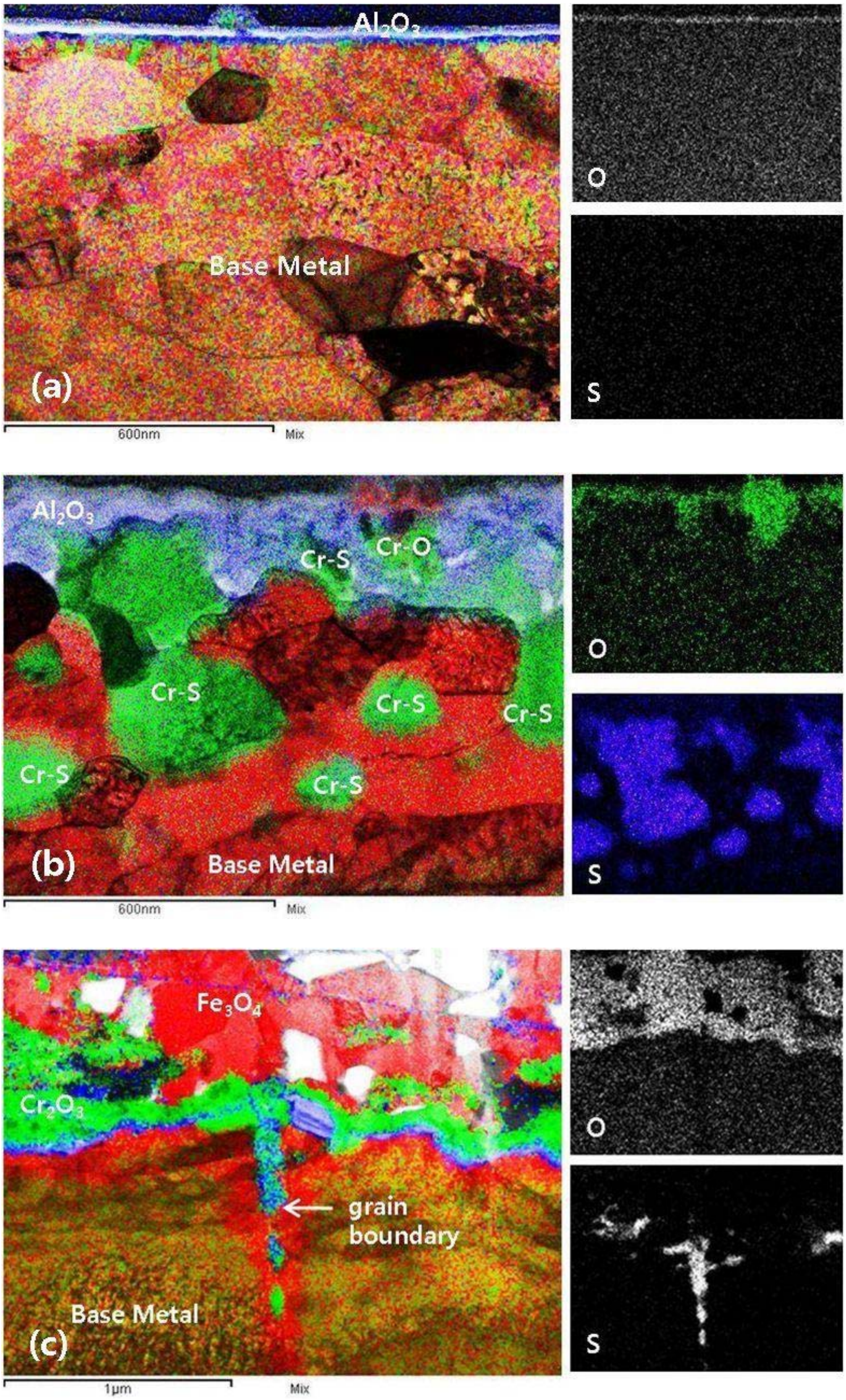
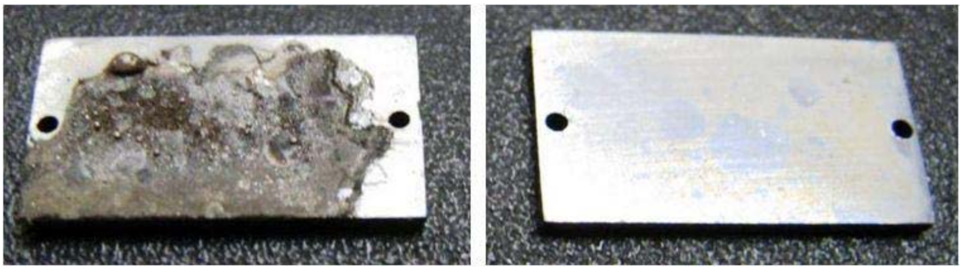


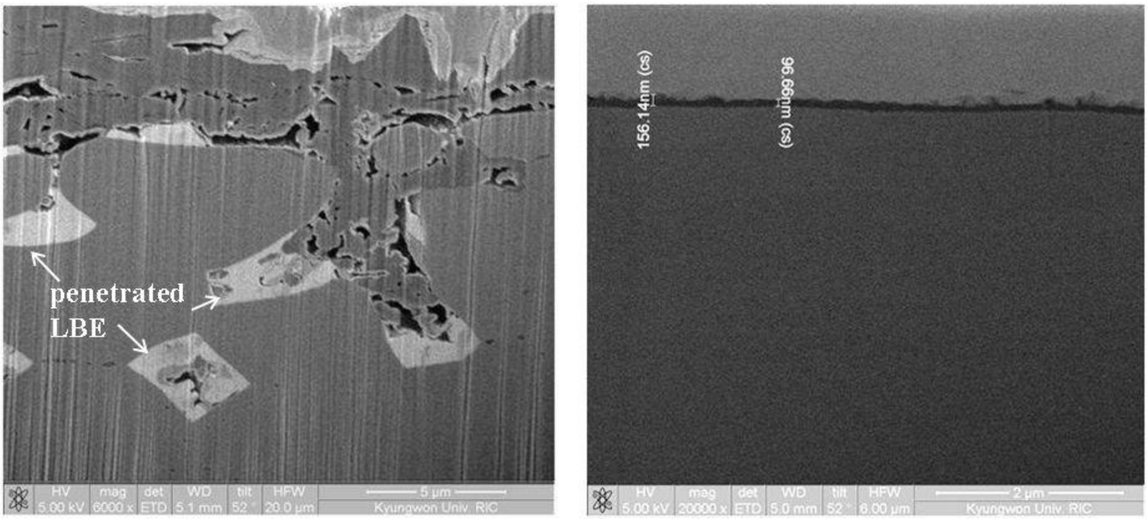
Figure 5: The photograph of surface of Kanthal®-AF after exposure to 650°C stagnant LBE for 500 hours



(a) As-received specimen

(b) H_2 heat-treated specimen

Figure 6: FESEM images of the cross-sectional area of Kanthal®-AF after exposure to 650°C stagnant LBE for 500 hours



(a) As-received specimen

(b) H_2 heat-treated specimen

Conclusion

Corrosion behaviors of iron-based alumina-forming alloys (Kanthal®-AF) were investigated in stagnant LBE at 500°C and 650°C for 500 hrs. From this study, the following conclusions are made:

- In 500°C tests, thin Al_2O_3 layers were formed on the surface of Kanthal®-AF and there was no dissolution attack by LBE. Co-segregation of S and Cr occurred at the oxide/substrate metal interface.
- Kanthal®-AF contains 40 ppm of sulphur. Even at such low levels of sulphur contents, sulphur was observed to segregate heavily in Al_2O_3/alloy interface, in the form of chromium sulphide. The formation of chromium sulphide is believed to promote the void formations at the oxide/alloy interface, and the voids consequently weaken the interfacial bonding and lead to severe dissolution attack in LBE.
- As the oxygen concentration at the scale region increases by inward diffusion of oxygen through the outer Al_2O_3 layer, the chromium sulphide transformed into Cr_2O_3. These processes may disturb the stable growth of Al_2O_3.
- The H_2 annealing heat treatment for reducing sulphur contents in Kanthal®-AF, significantly improves the protectiveness of oxide scale.

Acknowledgements

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Session V

Progress in transmutation physics experiments and nuclear data

Chairs: H. Oigawa, E.M. Gonzalez Romero

Nuclear data for advanced fuel cycles

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Abstract

A wide range of systems has been investigated, both within the AFCI and GNEP initiatives, in order to assess the impact of data uncertainty. In fact, some expected new significant features (core and fuel cycle) depend heavily on nuclear data knowledge and uncertainties. This paper reviews the results obtained, mostly in the framework of a NEA/NSC/WPEC Expert Group, and indicates directions for future work in order to improve TRU nuclear data.

1 Introduction

Within the Working Party on Evaluation Co-operation (WPEC) of the OECD Nuclear Energy Agency Nuclear Science Committee, a systematic approach has been developed to define data needs for advanced reactor and fuel cycle systems.

A comprehensive sensitivity and uncertainty study has been performed to evaluate the impact of neutron cross-section uncertainty on the most significant integral parameters related to the core and fuel cycle of a wide range of innovative systems.

These systems correspond to current studies in the Generation-IV initiative, the Global Nuclear Energy Partnership (GNEP), the Advanced Fuel Cycle Initiative (AFCI), and in advanced fuel cycle and partitioning/transmutation studies in Japan and Europe. In particular different types of Na-cooled fast reactors, a gas-cooled and a lead-cooled fast reactor, have been considered, as indicated in Table 1.

The integral parameters considered are related to the reactor core performances but also to some important fuel cycle-related parameters, like the transmutation potential, the doses in a waste repository or the neutron source at fuel fabrication [1].

Table 1: Features of the investigated systems

System	Fuel	Coolant	TRU/(U+TRU)	MA*/(U+TRU)	Power (MWth)
ABTR	Metal	Na	0.162	~ 0	250
SFR	Metal	Na	0.605	0.106	840
EFR	MOX	Na	0.237	0.012	3 600
GFR	Carbide	He	0.217	0.050	2 400
LFR	Metal	Pb	0.233	0.024	900
ADMAB	Nitride	Pb-Bi	1.0	0.680	380

* Minor actinides.

2 The covariance data

Since the uncertainty analysis depends very much on the assumed uncertainties on nuclear data, we show as examples some of the numerical values of the covariance data produced in the framework of Subgroup 26 activity [2]. In Table 2 diagonal values are shown for three typical minor actinides (*i.e.* ^{241}Am, ^{243}Am and ^{244}Cm).

Table 2: Diagonal values for three typical minor actinides

		^{241}Am						^{243}Am						^{244}Cm					
Gr	E [MeV]	ν	σ_f	σ_{inel}	σ_{el}	σ_{capt}	$\sigma_{n,2n}$	ν	σ_f	σ_{inel}	σ_{el}	σ_{capt}	$\sigma_{n,2n}$	ν	σ_f	σ_{inel}	σ_{el}	σ_{capt}	$\sigma_{n,2n}$
1	19.6	1.88	12.74	55.29	3.51	28.83	10.03	1.88	14.44	61.97	7.51	60.42	26.63	10.55	17.86	38.26	10.58	89.19	40.91
2	6.07	1.98	11.67	15.2	3.77	15.38	0	1.98	11.03	17.87	4.64	41.5	0	11.08	31.25	22.67	10.23	53.78	0
3	2.23	1.91	9.81	29.63	5.12	9.16	0	1.91	5.97	35.3	7.49	21.66	0	10.68	43.8	15.1	5.56	36.49	0
4	1.35	0.98	8.25	24.45	4.52	6.9	0	1.09	9.18	42.15	4.11	14.18	0	5.5	50.01	18.18	10.77	20.8	0
5	4.98e-1	1	8.29	23.03	5.5	5.29	0	1.2	9.62	40.98	5.9	8.92	0	5.6	36.53	29.09	9.33	22.54	0
6	1.83e-1	1	8.29	48.53	5.2	6.79	0	1.2	9.62	79.53	7.84	6.6	0	5.6	47.56	63.31	8.38	17.71	0
7	6.74e-2	1	7.39	51.78	4.81	7.96	0	1.2	7.12	80.77	4.41	4.57	0	5.6	26.26	59.72	9.21	17.43	0
8	2.48e-2	1	13.71	0	11.54	6.85	0	1.2	13.79	0	9.13	6.77	0	5.6	19.03	0	14.93	19.32	0
9	9.12e-3	1	13.51	0	12.35	6.66	0	1.2	13.54	0	9.6	6.64	0	5.6	11.92	0	14.04	12.14	0
10	2.03e-3	1	13.41	0	9.7	6.59	0	1.2	13.41	0	7.68	6.58	0	5.6	5.27	0	7.72	4.47	0
11	4.54e-4	1	8.08	0	14.53	3.67	0	1.2	9.64	0	8.96	2.31	0	5.6	5.7	0	3.61	4.6	0
12	2.26e-5	1	5.15	0	14.03	1.82	0	1.2	5.95	0	8.22	1.74	0	5.6	17.09	0	7.75	6.64	0
13	4.00e-6	1	6.72	0	14.2	5.54	0	1.2	4.81	0	7	3.43	0	5.6	21.99	0	6.62	11.79	0
14	5.40e-7	1	8.93	0	13.81	1.26	0	1.2	2.25	0	12.41	3.75	0	5.6	26.4	0	6.16	12.16	0
15	1.00e-7	1	3.02	0	13.03	1.8	0	1.2	2.12	0	11.44	3.58	0	5.6	27.18	0	6.12	12.51	0

These are preliminary data, and possible improvements are already foreseen. In Table 3, the ^{239}Pu and ^{241}Pu diagonal covariance data are shown for comparison. The ^{239}Pu fission uncertainty (*i.e.* a major potential contributor to the most important integral parameters uncertainty) is very small. On the contrary, the ^{241}Pu fission cross-section uncertainty is significant, and makes that isotope a major contributor to most integral parameter uncertainties.

Table 3: ^{239}Pu and ^{241}Pu diagonal covariance data

		^{239}Pu						^{241}Pu					
Gr	**E [MeV]**	ν	σ_f	σ_{inel}	σ_{el}	σ_{capt}	$\sigma_{n,2n}$	ν	σ_f	σ_{inel}	σ_{el}	σ_{capt}	$\sigma_{n,2n}$
1	19.6	**0.5**	0.63	23.06	6.94	37.08	8.53	0.45	24.09	25.15	4.45	55.39	39.68
2	6.07	0.17	0.69	22.18	9.36	37.8	4.34	0.27	14.16	19.47	3.74	54.1	33.43
3	2.23	0.17	0.89	19	10.3	26.56	0	0.27	21.26	18.38	4.39	38.41	0
4	1.35	0.12	0.64	29.01	10.29	18.18	0	0.28	16.62	19.78	5.38	31.66	0
5	4.98e-1	0.19	0.68	34.01	5.66	11.55	0	0.29	13.54	20.92	5.16	20.51	0
6	1.83e-1	0.54	0.85	46.06	3.98	9.04	0	0.29	19.87	30.09	4.69	11.29	0
7	6.74e-2	0.58	0.72	40.04	2.37	10.12	0	0.29	8.74	37.51	3.92	4.43	0
8	2.48e-2	0.58	0.96	28.52	2.16	7.39	0	0.29	11.29	0	9.14	7.79	0
9	9.12e-3	0.65	0.62	8.64	4.04	15.46	0	0.29	10.44	0	9.29	7.73	0
10	2.03e-3	**0.2**	1.2	0	0.74	1.39	0	0.29	12.68	0	10.96	7.74	0
11	4.54e-4	**0.2**	1.24	0	1.2	1.25	0	0.29	19.38	0	10.87	7.43	0
12	2.26e-5	**0.2**	0.47	0	0.24	0.61	0	0.29	4.21	0	10.66	8.38	0
13	4.00e-6	**0.2**	1.43	0	0.3	1.22	0	0.29	26.83	0	11.49	6.37	0
14	5.40e-7	**0.2**	0.88	0	0.44	1.36	0	0.29	2.94	0	9.91	6.84	0
15	1.00e-7	**0.2**	1.11	0	0.68	1.6	0	0.29	3.27	0	11.32	3.59	0

The process of improvement of the current evaluation of covariance data will be essential to consolidate their credibility and to define their field of application. At present, the results shown in the following paragraphs should be taken as meaningful orders of magnitude to be used in order to have guidelines and define priorities to improve the most important nuclear data, but they cannot be used yet directly, *e.g.* in a final design assessment. Moreover, with the consolidation of the covariance data, the best use of them will be in the frame of the so-called statistical adjustment/bias factor methods (see Section 8), where the integral experiment information will play an essential complementary role.

As for the impact of these covariance data, the calculated integral parameter uncertainties, resulting from the presently assessed uncertainties on nuclear data, are probably acceptable in the early phases of design feasibility studies [1]. In fact, the uncertainty on k_{eff} is less than 2% for all systems (with the exception of the accelerator-driven system, ADS) and reactivity coefficient uncertainties are below 20%. Power distributions uncertainties are also relatively small, except, once more, in the case of the ADS.

3 Target accuracies

Design target accuracies have also been agreed upon in the framework of Subgroup 26. Table 4 gives the design target accuracies presently adopted for fast neutron systems.

Table 4: Fast reactor and ADMAB target accuracies (1σ)

Multiplication factor (BOL)	300 pcm
Power peak (BOL)	2%
Burn-up reactivity swing	300 pcm
Reactivity coefficients (coolant void and Doppler at BOL)	7%
Major nuclide density at end of irradiation cycle	2%
Other nuclide density at end of irradiation cycle	10%

The unknown uncertainty data requirements can be obtained by solving a minimisation problem where the sensitivity coefficients in conjunction with the existing constraints provide the needed quantities to find the solutions:

$$\sum_i \lambda_i / b_i'^2 = \min$$

$$\sum_i S_{Ri} b_i'^2 S_{Ri}^+ + \sum_{i,j} S_{Ri} b_i' b_j' c_{ij} S_{Rj}^+ < Q_R$$

The c_{ij} are the correlation coefficients of the original data covariance matrix, and the b_i' are the unknown variance values needed to meet the requirements. To derive the expression above, the hypothesis has been made that the correlation terms in the covariance matrix do not change. The S_{Ri} are the sensitivity coefficients of integral parameter R to nuclear data i. Q_R is the target accuracy on the integral parameter R, and λ_i are cost parameters [3]. This expression can be easily generalised to a set of integral parameters and/or to a set of reference systems. To establish priorities in order to reduce uncertainties, the b_i' values should be compared to the variance data in the original covariance matrices. This analysis helps to decide if an action should be undertaken to meet the requirement.

Table 5 is a summary of the nuclear data target accuracies in the case of the full set of fast neutron systems analysed within Subgroup 26 [1]. These results show that, in the case of the wide range of fast reactors considered in this study, the priority requirement is for a drastic uncertainty reduction for some σ_{inel} (in particular for ^{238}U, but also for Fe and Na), for the σ_{fiss} of higher Pu isotopes and in particular for ^{241}Pu (between ~1-500 keV) and for σ_{capt} of ^{239}Pu (~1-500 keV).

Table 5: Summary of highest priority target accuracies for fast reactors

		Energy range	Current accuracy (%)	Target accuracy (%)
^{238}U	σ_{inel}	6.07 ÷ 0.498 MeV	10 ÷ 20	2 ÷ 3
	σ_{capt}	24.8 ÷ 2.04 keV	3 ÷ 9	1.5 ÷ 2
^{241}Pu	σ_{fiss}	1.35MeV ÷ 454 eV	8 ÷ 20	2 ÷ 3 (SFR, GFR, LFR) 5 ÷ 8 (ABTR, EFR)
^{239}Pu	σ_{capt}	498 ÷ 2.04 keV	7 ÷ 15	4 ÷ 7
^{240}Pu	σ_{fiss}	1.35 ÷ 0.498 MeV	6	1.5 ÷ 2
	ν	1.35 ÷ 0.498 MeV	4	1 ÷ 3
^{242}Pu	σ_{fiss}	2.23 ÷ 0.498 MeV	19 ÷ 21	3 ÷ 5
^{238}Pu	σ_{fiss}	1.35 ÷ 0.183 MeV	17	3 ÷ 5
^{242m}Am	σ_{fiss}	1.35MeV ÷ 67.4keV	17	3 ÷ 4
^{241}Am	σ_{fiss}	6.07 ÷ 2.23 MeV	12	3
^{244}Cm	σ_{fiss}	1.35 ÷ 0.498 MeV	50	5
^{245}Cm	σ_{fiss}	183 ÷ 67.4 keV	47	7
^{56}Fe	σ_{inel}	2.23 ÷ 0.498 MeV	16 ÷ 25	3 ÷ 6
^{23}Na	σ_{inel}	1.35 ÷ 0.498 MeV	28	4 ÷ 10
^{206}Pb	σ_{inel}	2.23 ÷ 1.35 MeV	14	3
^{207}Pb	σ_{inel}	1.35 ÷ 0.498 MeV	11	3
^{28}Si	σ_{inel}	6.07 ÷ 1.35 MeV	14 ÷ 50	3 ÷ 6
	σ_{capt}	19.6 ÷ 6.07 MeV	53	6

4 Impact of the minor actinide uncertainty data

For the purpose of assessing MA data needs, two systems (SFR and ADMAB) are of particular interest among the large number of systems investigated by Subgroup 26, since their fuel is loaded with large amounts of MA (see Table 6).

For these systems, the uncertainty due to nuclear data uncertainties of the most important integral parameters has been investigated; the results are summarised in Table 7.

Tables 8(a) and 8(b) show the major contributors to the uncertainty, both Pu isotopes and selected minor actinides.

The impact of nuclear data uncertainties on the nuclide density variations $\Delta n = n_f - n_0$ during irradiation are shown in Tables 9(a) and 9(b), where n_f is the density at the end of irradiation and n_0 the density at beginning of irradiation, for both the SFR and the ADMAB systems.

Table 6: Some features of the SFR and ADMAB systems

System	Fuel	Coolant	TRU/(U+TRU)	MA/(U+TRU)	Power (MWth)
SFR	Metal	Na	0.605	0.106	840
ADMAB-ADS	Nitride	Pb-Bi	1.0	0.680	380

Table 7: Fast neutron systems with high MA content – total uncertainties (%)

Reactor	k_{eff}	Power peak	Doppler	Void	Burn-up [pcm]	Decay heat	Dose	Neutron source
SFR	**1.82**	0.4	5.6	**17.1**	272	0.4	0.3	1.0
ADMAB	**2.94**	**21.4**	–	**15.5**	**1 044**	0.7	1.0	2.5

Table 8(a): Uncertainties (%) due to selected MA data

		SFR		ADMAB		
Isotope	Cross-section	k_{eff}	Void	k_{eff}	Power peak	Void
^{241}Am	Fission	0.08	0.4	0.83	5.8	3.3
^{242m}Am	Fission	0.73	3.7	0.14	1.1	0.3
^{243}Am	Fission	0.04	0.3	0.35	2.4	1.6
^{244}Cm	Fission	0.39	3.0	1.90	13.4	3.2
^{245}Cm	Fission	0.39	1.0	1.04	7.6	1.6

Table 8(b): Uncertainties (%) due to Pu isotope data

		SFR		ADMAB		
Isotope	Cross-section	k_{eff}	Void	k_{eff}	Power peak	Void
^{238}Pu	ν	0.34	0.4	0.13	0.9	0.2
	Fission	0.53	2.9	0.21	1.5	0.4
^{239}Pu	Fission	0.12	0.9	0.12	0.9	0.3
	Capture	0.12	1.2	0.10	0.8	0.5
^{240}Pu	ν	0.39	2.2	0.14	1.0	0.2
	Fission	0.44	2.6	0.16	1.2	0.2
	Capture	0.31	1.8	0.08	0.6	0.3
^{241}Pu	Fission	0.96	4.1	1.04	7.6	2.0
^{242}Pu	Fission	0.36	2.5	0.15	1.1	0.3
	Capture	0.17	2.2	0.06	0.4	0.3

Table 9(a): ADMAB – Δn uncertainty (%)

		^{241}Pu	^{242}Pu	**^{241}Am**	**^{242m}Am**	**^{243}Am**	**^{242}Cm**	**^{244}Cm**	**^{245}Cm**	**^{246}Cm**
^{241}Pu	Capture	2.18	4.42	0.01	–	–	–	–	–	–
	Fission	9.18	0.27	0.05	–	–	–	–	–	–
^{242}Pu	Capture	–	5.80	–	–	1.03	–	–	–	–
	Fission	–	3.72	–	–	0.01	–	–	–	–
^{241}Am	**Capture**	–	5.05	4.18	5.92	–	4.73	–	–	–
	Fission	–	0.09	1.51	0.11	–	0.11	–	–	–
^{242m}Am	**Capture**	–	–	–	0.80	–	–	–	–	–
	Fission	–	–	–	3.81	–	–	–	–	–
^{243}Am	**Capture**	–	–	–	–	4.27	–	9.40	1.93	0.03
	Fission	–	–	–	–	1.17	–	0.11	0.02	–
^{242}Cm	**Fission**	–	–	–	–	–	0.54	–	–	–
^{244}Cm	**Capture**	–	–	–	–	–	–	3.93	57.10	1.38
	Fission	–	–	–	–	–	–	15.13	2.96	0.05
^{245}Cm	**Capture**	–	–	–	–	–	–	–	3.78	12.08
	Fission	–	–	–	–	–	–	–	90.97	2.21
^{246}Cm	**Fission**	–	–	–	–	–	–	–	–	1.39
Total		9.97	9.62	**4.45**	**7.09**	**4.54**	**4.76**	**18.24**	**107.52**	**12.44**

Table 9(b): SFR – Δn uncertainty (%)

		^{241}Pu	^{242}Pu	**^{241}Am**	**^{242m}Am**	**^{243}Am**	**^{242}Cm**	**^{244}Cm**	**^{245}Cm**	**^{246}Cm**
^{241}Pu	Capture	2.77	4.23	0.04	–	–	0.01	–	–	–
	Fission	19.38	0.21	0.31	–	–	0.06	–	–	–
^{242}Pu	Capture	–	12.83	–	–	49.38	–	3.23	–	–
	Fission	–	15.25	–	–	0.37	–	0.02	–	–
^{241}Am	**Capture**	–	0.69	5.41	0.44	–	44.66	–	–	–
	Fission	–	0.01	2.99	0.01	–	0.84	–	–	–
^{242m}Am	**Capture**	–	–	–	2.15	14.13	0.04	–	–	–
	Fission	–	–	–	10.65	0.58	0.22	–	–	–
^{243}Am	**Capture**	–	–	–	–	14.46	–	38.66	0.21	–
	Fission	–	–	–	–	6.52	–	0.43	–	–
^{242}Cm	**Fission**	–	–	–	–	–	15.02	–	–	–
^{244}Cm	**Capture**	–	–	–	–	–	–	28.94	18.11	–
	Fission	–	–	–	–	–	–	145.34	0.77	–
^{245}Cm	**Capture**	–	–	–	–	–	–	–	2.81	18.13
	Fission	–	–	–	–	–	–	–	61.89	2.19
^{246}Cm	**Capture**	–	–	–	–	–	–	–	–	24.45
	Fission	–	–	–	–	–	–	–	–	51.16
Total		24.61	20.39	**6.20**	**10.88**	**53.76**	**47.18**	**153.19**	**64.55**	**59.57**

5 MA neutron cross-section target accuracy requirements

Using the approach outlined in Section 3, the requirements listed in Tables 10(a) and 10(b) are found for MA data.

These results show that when the fuel is heavily loaded with MA tight requirements are found for some MA cross-sections, in particular for σ_{fiss} of ^{244}Cm, ^{241}Am, ^{245}Cm, ^{243}Am, ^{242}Cm, ^{242m}Am, for σ_{inel} of ^{243}Am and for ν of ^{244}Cm. For these reactions, the required accuracies are an order of magnitude below the present uncertainties. As for the major actinides, it is confirmed that, also for these systems, improvements are required for the σ_{fiss} of ^{241}Pu (again ~ factor 10), for the σ_{fiss} of ^{238}Pu (~ factor 5) and for ν of ^{238}Pu (~ factor 3).

Table 10(a): ADMAB – MA uncertainty reduction required to meet design target accuracies

Isotope	Cross-section	Energy range	Uncertainty (%)	
			Initial	Required
^{244}Cm	σ_{fiss}	6.07-2.23 MeV	31.3	3.0
		2.23-1.35 MeV	43.8	2.6
		1.35-0.498 MeV	50.0	1.5
^{243}Am	σ_{inel}	1.35-0.498 MeV	42.2	2.3
		498-183 keV	41.0	3.6
		183-67.4 keV	79.5	3.7
^{241}Am	σ_{fiss}	6.07-2.23 MeV	11.7	1.7
		2.23-1.35 MeV	9.8	1.4
		1.35-0.498 MeV	8.3	1.2
^{245}Cm	σ_{fiss}	1.35-0.498 MeV	49.4	3.3
		498-183 keV	37.2	2.9
		183-67.4 keV	47.5	2.9
		67.4-24.8 keV	26.5	3.2
^{243}Am	σ_{fiss}	6.07-2.23 MeV	11.0	2.3
		1.35-0.498 MeV	9.2	1.6
^{244}Cm	ν	6.07-2.23 MeV	11.1	2.5
		1.35-0.498 MeV	5.5	1.3
^{242}Cm	σ_{fiss}	6.07-2.23 MeV	52.6	26
		498-183 keV	66.0	28.4
^{242m}Am	σ_{fiss}	498-183 keV	16.6	4.8
		183-67.4 keV	16.6	4.8

Table 10(b): SFR – MA uncertainty reduction required to meet design target accuracies

Isotope	Cross-section	Energy range	Uncertainty (%)	
			Initial	Required
^{244}Cm	σ_{fiss}	1.35-0.498 MeV	50.0	5.1
^{242m}Am	σ_{fiss}	1.35-0.498 MeV	16.5	4.2
		498-183 keV	16.6	3.1
		183-67.4 keV	16.6	3.1
		67.4-24.8 keV	14.4	4.0
		24.8-9.12 keV	11.8	4.2
		2.04-0.454 keV	12.2	5.1
		1.35-0.498 MeV	19.0	3.5
^{245}Cm	σ_{fiss}	183-67.4 keV	47.5	6.7

6 Other data needs

The previous discussion is focused on “neutron cross-section” data needs. Some fuel-cycle-related data needs are implicitly accounted for since nuclide densities at the end of irradiation were included in the analysis. However, fuel cycle data needs should be specified for the very specific fuel cycle(s) to be associated to a dedicated minor actinide burner, as foreseen, *e.g.* in the “double strata” fuel cycle approach. In this respect, a specific analysis should be made on the decay heat data needs (both in the reactor and at fuel discharge/storage) when a fuel highly loaded (*e.g.* ~50% HM) is considered. The relative contribution of heavy isotopes (and its breakdown by isotope) and fission products at different cooling times of the decay heat both in a dedicated burner reactor with a highly MA loaded fuel and in Superphénix are shown in Tables 11(a) and 11(b) [4]. The data show the importance of the MA contribution and, as a consequence, they underline the need for high accuracy data.

Finally, it has been indicated in Subgroup 26 the need to evaluate in detail the impact of uncertainties on the fission spectrum of the isotopes that contribute the most to the fission source in the reactor. This can be the case of MA in dedicated MA burners.

Table 11(a): Decay heat – relative contribution of heavy isotopes and fission products at different cooling times*

	Discharge[a]	500 s	1 000 s	3 000 s	1 h	12 h	1 day	10 days
ADS								
Heavy elements	23	46	50	57	58	74	77	86
Fission products	77	53	50	43	41	26	22	14
Superphénix								
Heavy elements	8.9	NA	20.2	22.3	22.5	32.3	34.5	22.8
Fission products	89.7	NA	74.6	72.6	72.3	63.7	62.1	73.2

* Relative contribution (%).

[a] EOL (2 yr).

Figure 11(b): Decay heat – heavy element breakdown by isotope*

	Discharge[a]	500 s	1 000 s	3 000 s	1 h	12 h	1 day	10 days
U	7.63E+0[b]	7.62E+0	7.61E+0	7.59E+0	7.58E+0	7.29E+0	7.01E+0	3.71E+0
Np	3.05E+5	3.04E+5	3.04E+5	3.01E+5	3.01E+5	2.58E+5	2.19E+5	1.15E+4
Pu	9.59E+4	9.58E+4	9.56E+4	9.50E+4	9.49E+4	8.93E+4	8.81E+4	8.85E+4
Am	9.08E+5	7.73E+5	6.65E+5	4.08E+5	3.66E+5	1.73E+5	1.34E+5	7.83E+4
Cm	4.33E+6	4.33E+6	4.33E+6	4.33E+6	4.33E+6	4.33E+6	4.33E+6	4.20E+6
Bk	1.37E–3	1.35E–0	1.33E–3	1.26E–3	1.26E–3	7.09E–4	6.58E–4	6.41E–4
Cf	2.16E–4	2.16E–4	2.16E–4	2.16E–4	2.16E–4	2.17E–4	2.17E–4	2.22E–4
Total	5.64E+6	5.51E+6	5.40E+6	5.14E+6	5.09E+6	4.85E+6	4.77E+6	4.38E+6

* Decay heat (W).

[a] EOL (2 yr).

[b] Read as 7.63×10^0.

7 How to meet requirements

The results shown in Sections 3 and 5 indicate some very tight requirements for some Pu and MA isotopes (^{244}Cm, ^{243}Am, etc.), essentially for the fission cross-section data. In order to meet these requirements and to obtain such tight accuracies, it is generally considered to be very important to perform appropriate integral experiments, due to potential difficulties associated with differential measurements. However, even the integral experiments available are relatively limited, and some of the highest accuracy integral experiments give information essentially on capture cross-sections. A short discussion is given below.

PROFIL-type experiments in PHENIX

Such experiments consist in irradiating small samples of separated isotopes in well-characterised spectral conditions

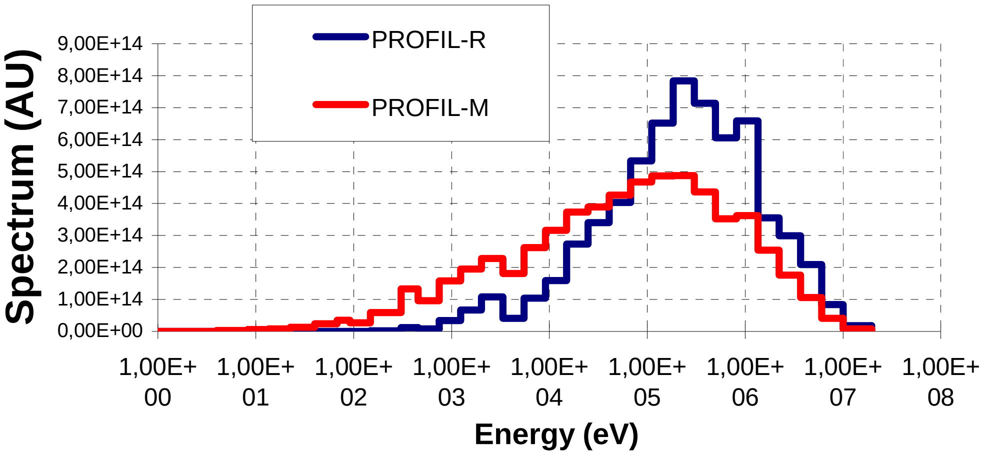
After irradiation, the contents of the sample containers are dissolved and analysed by mass spectroscopy. Un-irradiated samples are similarly analysed.

The difference in contents between the irradiated and un-irradiated samples provides "direct" information on isotopic capture reactions.

PROFIL experiments have been heavily used in nuclear data adjustment studies, both in France [5] and in the USA. Some data related to Am isotopes have provided preliminary information on their capture data and, for a specific case, on a branching ratio.

A new series of experiments is under way in PHENIX: PROFIL-R and -M (*i.e.* with or with or without a moderator surrounding the samples, to soften the spectrum), with a larger number of MA samples. The PROFIL neutron spectra are compared in Figure 1.

Figure 1: Neutron spectra in the PROFIL-R and PROFIL-M subassemblies



Fission rate measurements in critical assemblies

As for fission, a few experiments have been performed in the MASURCA critical facility with MA fission chambers (*e.g.* during the MUSE programme).

These experiments could be repeated with a larger number of MA fission chambers, and using variable moderators surrounding the fission chamber, to gain information on the fission cross-section over a wide energy range.

Other critical experiments

Finally, a systematic integral experiment programme has been performed at the FCA critical facility in the past, aiming to the reactivity assessment of selected MA in variable spectrum environments.

8 Combined use of covariance data and integral experiments

Methodologies for reducing uncertainties coming from nuclear data on the main fast reactor neutronic design parameters rely on the use of integral experiment information in a statistical multi-group cross-section adjustment.

A combined use of scientifically-based covariance data and of selected integral experiments can be made using classical statistical adjustment techniques [6,7]. These techniques provide adjusted nuclear data for a wide range of applications, together with new, improved covariance data and bias factors (with reduced uncertainties) for the required design parameters, in order to meet target accuracies.

A preliminary study has been performed [8], combining covariance data and integral experiments to statistically adjust the cross-section data in order to meet design target accuracies of the type shown previously. The results are encouraging, and it has been suggested to perform more complete studies in that field, within initiatives of the OECD/NEA/NSC [9].

9 Conclusions

Nuclear data requirements for innovative systems have been systematically assessed, using sensitivity and uncertainty analysis, and preliminary covariance data. This work was performed mostly in the framework of NEA Nuclear Science Committee activities. The approach to meet these requirements needs the use of differential and integral experiments, to be combined within a statistical adjustment method. This area should also be explored within an international collaboration, if launched in a timely way. This approach will allow the improvement of nuclear data, in particular in the field of inelastic cross-sections, MA and Pu isotope fission cross-sections.

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Cross-section measurements of minor actinides at the n_TOF-Ph2 experiment at CERN

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Abstract

The n_TOF facility is a spallation neutron source driven by the CERN proton synchrotron. It is coupled to a 200 m long flight path and used for neutron cross-section measurements. The facility was designed, built and commissioned between 1999 and 2001 and operated between 2001 and 2004. The n_TOF collaboration has successfully completed an ambitious experimental programme on neutron cross-section measurements of isotopes relevant to the transmutation of nuclear waste and advanced nuclear technologies. A new experimental programme n_TOF-Ph2 will be started in 2008, after the installation and commissioning of a new spallation target.

Combined neutron capture and neutron-induced fission measurements of various fissile minor actinides (MA) will be performed with especially designed active targets: the capture and fission yields will be obtained simultaneously using the n_TOF Total Absorption Calorimeter (TAC) and mini-ionisation chambers containing the isotopically enriched metallic samples, respectively. The capture cross-section measurements of non-fissile MA will be performed only with the TAC.

Introduction

The increase of the world energy demand and the need of low carbon energy sources have triggered the renaissance and/or enhancement of nuclear energy in many countries. Fundamental nuclear physics can contribute in a practical way to the sustainability and safety of the nuclear energy production and the management of the nuclear waste. There exists a series of recent studies which address the most relevant isotopes, decay data, nuclear reaction channels and energy ranges which must be investigated in more detail for improving the design of different advanced nuclear systems [35] and nuclear fuel cycles [36].

New concepts for nuclear systems are being explored to improve the sustainability of nuclear energy that appears in the EU SET plan [37] as an unavoidable component in the mix for energy generation. These new concepts cover notions from evolutionary light water reactors, which will most probably remain the main component of the nuclear park during this century, to fast reactors foreseen in Generation IV, as the only really long-term sustainable systems by optimising the natural resources, and include subcritical fast systems, ADS, especially suited for intensive waste minimisation.

Despite the many previous measurements and recent efforts, the optimisation and future industrial deployment of these new concepts still challenge the present level of basic nuclear data knowledge. The availability of all the required data, their quality and the description of their uncertainties in the most recent databases and simulation tools need to be improved. The main challenges come from:

- Isotopes which have become relevant due to their increased presence in the fuels for the new generation of reactors. This applies in particular to the systems aiming at the Pu and minor actinide (MA) recycling: fast reactors and ADS, but also Gen-III(+) reactors with a high Pu load.
- The proposal of fast systems, which has enhanced the relevance of the region from 1 eV to several MeV for all isotopes and, very especially, for high mass actinides.
- New fuel cycles with multi-recycling of actinides that could drive to new levels of accumulation of minority isotopes, thus generating additional risks and/or costs at several stages of the fuel cycle. One of the most important aspects is the propagation of the uncertainties as the number of irradiation cycles increases.
- New requirements on the level of precision. In the nuclear industry as everywhere else, the computer simulations should help minimising (but not replacing) the need for small scale and costly experimental demonstrators. This new role of the simulation tools will require a higher level of precision that can only be achieved if the precision of initial basic nuclear data is previously improved.
- Better assessment of the uncertainties on the data and on the derived magnitudes. This requires a more realistic evaluation of the cross-section uncertainties and their correlations between different energies and different channels. Only in this way will the estimations from the simulations profit from the enhanced basic data precision and from the constraints set by the integral experiments.

As a consequence, new data measurements, evaluations of cross-sections with reliable covariance data, and integral experiment validations are needed for ADS systems in addition to the other fast reactor system requirements. Linear optimisation systems had been used to get recommendations on possible priorities to improve the present situation with specific improvement on selected nuclear data. However these priorities have to be additionally weighted taking into account the experimental difficulties and resources requirements in order to establish the practical priorities for new data needs.

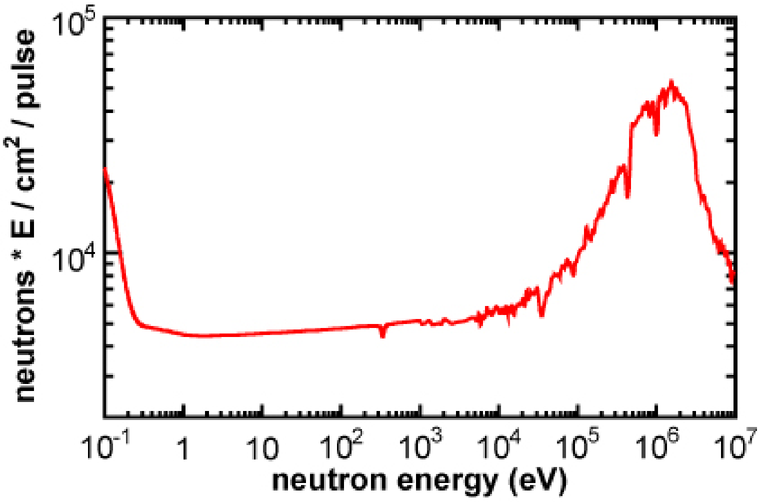
The n_TOF facility at CERN was built in 2000 with the goal of providing the international nuclear data community with a unique place to measure neutron-induced cross-sections of highly radioactive isotopes available only in small amount. Since then, it has made essential contributions to the field of the nuclear data for the science and the technology and, in particular, has produced the best data sets on the neutron capture cross-section of ^{237}Np and ^{240}Pu [15].

The n_TOF facility

A new neutron time-of-flight facility was built at CERN in 2001 as the result an idea proposed by Rubbia, *et al.* [9]. The n_TOF facility [10] has been exploited scientifically during the period between 2001 and 2004 by an international collaboration. The neutrons are produced by spallation reactions induced by the CERN PS proton beam with 6 ns width, 20 GeV/c and up to 7 × 1 012 protons. Each pulse impinges on a 80 × 80 × 60 cm³ lead target and yields about 300 neutrons per proton. A 5 cm thick water pool is used as both as coolant of the lead target and as a moderator of the spallation neutron spectrum. The resulting neutron energy spectrum ranges from 1 eV to 250 MeV with a nearly 1/E isolethargic flux dependence up to 1 MeV. The neutrons travel to the experimental area at a 185 m flight distance inside a vacuum pipe. A sweeping magnet with 1.5 T has been placed at a distance of 145 m of the spallation target for deflecting the accompanying charged particles. They are stopped by a thick iron shielding placed 2 meters after the magnet.

Two collimators consisting of layers of iron and borated polyethylene are used for collimating and shaping the neutron beam. The iron is used for scattering away the neutrons and the borated polyethylene moderates and absorbs the neutrons. The first collimator has an inner diameter of 11 cm, an outer diameter of 50 cm and is placed 135 m from the lead target. The second collimator is located near the experimental area at a distance of 175 m and has an outer diameter of 40 cm. Neutron beams for neutron capture measurements are shaped with an inner diameter of 1.8 cm. The energy distribution and magnitude of the neutron flux has been measured with a ^{235}U fission chamber calibrated at the Physikalisch Technische Bundesanstalt in Braunschweig [11] and is shown in Figure 1. The broader neutron beams for fission measurements are shaped with an alternative second collimator of 8 cm inner diameter. The neutron beams are Gaussian-shaped beam profile at the position where the samples are placed.

Figure 1: Shape and magnitude of the neutron flux at 185 m from the spallation target



Experimental techniques

The n_TOF collaboration has developed several types of detectors for both capture and fission cross-section measurements.

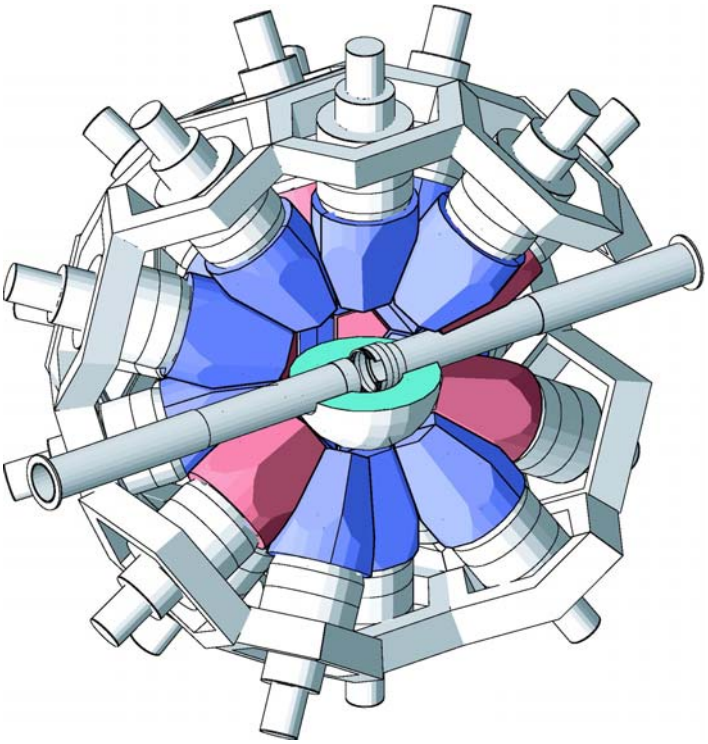
Neutron capture (n,γ)

Two neutron cross-section measurement techniques have been applied at n_TOF. The first is based on deuterated benzene C_6D_6 total energy gamma-ray detectors contained in a cylindrical low mass carbon fibre housing [13]. The samples are placed in the beam by a carbon fibre sample changer. The low neutron capture cross-sections of both the carbon fibre and the benzene assure a low neutron sensitivity, *i.e.* neutrons scattered at the sample and detected as gamma-ray events.

The total energy detectors have a cascade detection efficiency which is nearly proportional to the capture cascade energy. Accurate proportionality is achieved by the use of weighting functions computed by Monte Carlo simulations [14]. The detection efficiency of a capture event for a two C6D6 set-up is 20%.

Cross-sections of highly radioactive samples, usually available in small amounts, are measured with the total absorption spectroscopy technique. The n_TOF collaboration built in 2004 [15] a 4π total absorption calorimeter (TAC) made up of 40 BaF_2 crystals (Figure 2). The crystals are embedded in 10B doped carbon fibre capsules and coupled to XP4512B photomultipliers. The TAC has a 100% rate of efficiency in detecting capture events. The radioactive samples are placed at the geometric centre of the TAC and are surrounded by a $C_{12}H_{20}O_4(^6Li)_2$ neutron absorber, for moderating and absorbing the scattered neutrons.

Figure 2: Total absorption calorimeter consisting of 40 BaF_2 crystals



All neutron capture cross-sections area measured relative to ^{197}Au(n,γ) which is a standard cross-section in the energy region above 200 keV and has a well measured saturated resonance at 4.9 eV.

The relative variations of the integrated neutron flux, *i.e.* the shape of the neutron kinetic energy distribution of the flux, are measured continuously during the experiments with a monitor based on the ^{6}Li(n,α) reaction [16]. In addition, three $^{10}BF_3$ detectors are installed in the neutron escape lane for monitoring purposes.

Neutron-induced fission (n,f)

Fission cross-section measurements have been carried out with two independent detector systems.

Two Fast Induction Chambers (FIC) have been developed and are used at the n TOF facility. The detector consists of a stack of several fissile isotopes with a thickness of 200 μg/cm^2 deposited on 100 μm thick aluminium foils. The first FIC-0 detector was loaded with low activity samples and the second FIC-1 detector was used for very radioactive samples with higher activity [17].

The second fission set-up consists of a stack of Parallel Plate Avalanche Counters (PPAC). The fission samples were deposited on 1.5 μm thick Mylar or 2 μm aluminium foils and placed in the middle of two PPAC. The system allows to measure several samples on the row and to reconstruct the interaction point by measuring in coincidence the trajectories of the two fission fragments [18].

The fission cross-sections are measured relative to the ^{235}U(n,f) standard and to ^{238}U(n,f).

The data acquisition system

The n_TOF collaboration has dedicated a major effort to developing a pioneering and fully digital data acquisition system (DAQ) [19]. The DAQ is based on the sampling of the detector signals in order to extract the deposited energy and the time of flight. A grand total of 54 Acqiris flash ADC with 8-bit amplitude resolution and 1 ns sampling interval with 8 Mbytes of memory was used to record the full detector signal following the start time given by the incident protons. The digitisers were operated at 500 Msamples/s, allowing to store the detector signal during 16 ms, corresponding to a time-of-flight of 0.3 eV.

The digital data are zero suppressed and transferred to CERN's Central Data Recording. The raw data are processed and reduced to summary tapes by pulse shape analysis software running on batch at CERN's LXBATCH facility.

Cross-section measurements

During the first phase of data-taking two types of cross-section experiments were performed, neutron capture gamma cross-section measurements with the C6D6 and BaF_2 detectors, and neutron-induced fission measurements with the FIC and PPAC detectors. A summary of these measurements is given in Table 1.

The neutron capture measurements of ^{24,25,26}Mg, ^{56}Fe, ^{90,91,92,93,94,96}Zr [20], ^{139}La [21], ^{151}Sm [22,23], ^{186,187,188}Os [24], ^{197}Au [25], ^{204}Pb [26], ^{206}Pb [27], ^{207}Pb [28], ^{208}Pb [29], ^{209}Bi [30,31] and ^{232}Th [32] were performed with the C_6D_6 detectors. The BaF2 total absorption calorimeter has been used for measurements of ^{197}Au [25], ^{233}U [32], ^{234}U [33], and ^{237}Np, ^{240}Pu, and ^{243}Am [34]. An additional test measurement of the fissile isotope ^{233}U was performed with the TAC as a preparatory test for the n_TOF-Ph2 experiments.

Table 1: Cross-sections measurements done in the period 2002-2004

Technique and detector	Isotopes
TAC (n,γ)	^{197}Au, ^{233,234}U, ^{237}Np, ^{240}Pu, ^{243}Am
C_6D_6 (n, γ)	^{24,25,26}Mg, ^{56}Fe, ^{90,91,92,93,94,96}Zr, ^{139}La, ^{151}Sm, ^{186,187,188}Os, ^{197}Au, ^{204,206,207,208}Pb, ^{209}Bi, ^{232}Th
PPAC (n,f)	^{nat}Pb, ^{209}Bi, ^{232}Th, ^{237}Np, ^{233,234,235,238}U
FIC (n,f)	^{232}Th, ^{237}Np, ^{233,234,235,236,238}U, ^{241,243}Am, ^{245}Cm

For the capture measurements, the sample masses were relatively high (~1 g) except for the very radioactive ones (~10-100 mg). The measurements were performed with only one sample in the beam. The fission experiments allowed measuring various samples during the same beam time. This allowed an efficient use of the neutron beam, since the samples for fission should not exceed a surface density of a few hundred μg/cm^2 for minimising the self absorption of the fission fragments.

The fission cross-sections measured with the FIC-0 detector correspond to ^{232}Th, ^{234}U, ^{235}U, ^{236}U, ^{238}U and ^{237}Np isotopes. The isotopes measured with FIC-1were ^{233,235,238}U, ^{241}Am, ^{243}Am and ^{245}Cm. The fission cross-sections of ^{nat}Pb, ^{209}Bi, ^{232}Th, ^{237}Np, ^{233,234,235,238}U have been measured with Parallel Plate Avalanche Counters (PPAC) [17,18].

The second phase of n_TOF: n_TOF-Ph2

The n_TOF facility will restart operation in 2008. Table 2 shows the measurements proposed for the second phase of the experiment n_TOF-Ph2.

Table 2: Cross-sections measurements proposed for the n_TOF-Ph2

Cross-section	Isotopes
(n,γ)	Mo, Ru Pd stable isotopes Fe, Ni, Zn, Se stable isotopes Various A~150 isotopes ^{234,235,236,238}U, ^{231,233}Pa ^{239,240,242}Pu, ^{241,243}Am, ^{245}Cm
(n,f)	^{231}Pa, ^{244,245}Cm, ^{241}Pu, ^{241,243}Am, ^{234,235}U
(n,α)	^{147}Sm, ^{67}Zn, ^{99}Ru

The experimental programme follows the lines defined during the first three experimental campaigns in 2002, 2003 and 2004, which identified the three main objectives of the experimental activities at n_TOF: nuclear data measurements for advanced nuclear technologies and nuclear waste transmutation, neutron cross-section measurements for nuclear astrophysics and neutron cross-section measurements for basic nuclear physics.

Summary and conclusions

The first phase of data-taking of the n_TOF facility (2002-2004) has been a success. The installation has not been running in the period 2005-2007 due to the PS shutdown for the Large Hadron Collider construction and an upgrade of the infrastructure of the facility is expected in 2007.

The cross-sections are measured by the time-of-flight method over a broad neutron energy range. The high instantaneous flux at the n_TOF facility allows measuring highly radioactive samples available only in low masses.

A new series of capture and fission measurements is planned for n_TOF-Ph2, a second phase of operation. It is foreseen to perform simultaneous fission and capture cross-section measurements on fissile isotopes.

An upgrade to the facility with a second flight path at a shorter distance of 20 m is under discussion for the third n_TOF phase. Such a short flight path will offer a neutron beam with 100 times more neutron flux and would therefore make possible measurements which are not feasible nowadays.

Acknowledgements

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Status of J-PARC Transmutation Experimental Facility

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Abstract

Construction of the Transmutation Experimental Facility (TEF) is planned within the framework of the Japan Proton Accelerator Research Complex (J-PARC) project. The facility has a critical assembly and a lead-bismuth target connected to a 600 MeV proton beam line. In the current J-PARC schedule, it is planned to install the critical assembly with a 30 kW beam dump during the first phase and the lead-bismuth target facility with superconducting linac will be built at the latter phase. Based on this construction plan, Japan Atomic Energy Agency (JAEA) collected the preliminary Letters Of Intent (LOI) for TEF. About 40 research programmes from a hundred researchers were proposed from all over the world. The proposals spread over wide research fields such as innovative reactor concepts including ADS and FBR, nuclear data, nuclear physics and so on. A medical application using 30 kW proton beam dump was also proposed. According to these many valuable proposals, the rearrangement of the facility layout and the new experimental equipment design are now under way. One of the important developments is to prepare for the application of minor actinide fuels at critical assembly. The development of the technology to extract low-power proton beam, handling method for minor actinide fuel is being performed. A sensitivity analysis was also performed to determine the efficient fuel composition and experimental method. The latest facility concept reflecting these R&D efforts and user requests based on LOI will be introduced.

Introduction

The Japan Atomic Energy Agency (JAEA) is performing a design study of the accelerator-driven system (ADS) to transmute minor actinides and long-lived fission products [1]. To study the basic characteristics of ADS and to demonstrate its feasibility from the viewpoints of reactor physics and spallation target engineering, JAEA plans to build the Transmutation Experimental Facility (TEF) at the JAEA Tokai site within the framework of the J-PARC Project, which is jointly directed by JAEA and the High Energy Accelerator Research Organization (KEK) [2]. Figure 1 illustrates the site plan of J-PARC. As shown in Figure 2, the TEF consists of two buildings; Transmutation Physics Experimental Facility (TEF-P) [3] and ADS Target Test Facility (TEF-T) [4]. TEF-P is a zero-power critical facility where a low-power proton beam is available to research the reactor physics and the controllability of the ADS. It also has an availability to measure the reaction cross-sections of MA, structural materials and so on. TEF-T is planned as a material irradiation facility which can accept a maximum 600 MeV-200 kW proton beam into the Pb-Bi eutectic alloy spallation target. Using these two facilities, the basic physical properties of subcritical system and engineering tests of spallation target will be studied.

Figure 1: Site plan of J-PARC Project

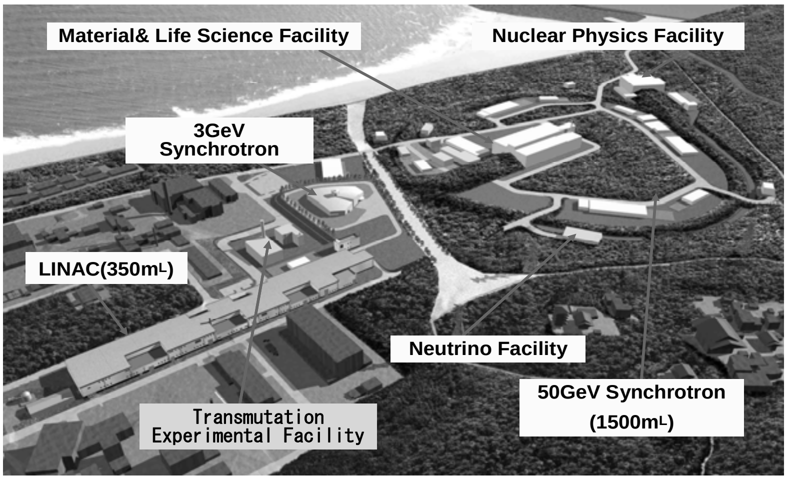
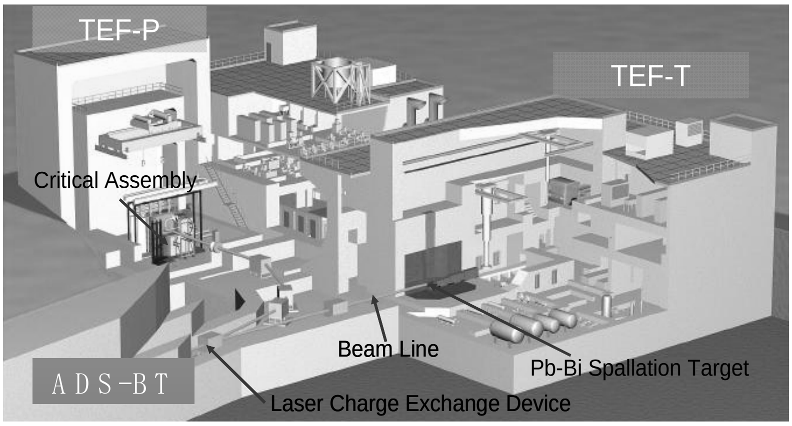


Figure 2: Transmutation Experimental Facility



Research and development (R&D) for several important technologies required to build the facilities are also performed, such as a laser charge exchange technique to extract a very low-power beam for reactor physics experiments, a remote handling method to load minor actinide (MA) bearing fuel into the critical assembly and so on. The objectives of the facilities, the latest design concept and key technologies to construct TEF are described.

Outline of TEF-P

Several ADS neutronics experiments have been performed, such as the MUSE programme [5] at the MASURCA critical assembly in France using a DT and DD neutron source and a solid target. In Japan, subcritical experiments were performed at the Fast Critical Assembly (FCA) at JAEA Tokai with a ^{252}Cf neutron source and a DT neutron source. Subcritical experiments with thermal subcritical core driven by DT neutron and medium-energy proton are now under way at Kyoto University Research Reactor Institute. Experimental studies have also been performed concerning the neutronics of the spallation neutron source with various target materials such as lead, tungsten, mercury and uranium. These experiments for spallation target are also useful to validate the neutronics characteristics of ADS.

There have been, however, no subcritical experiments combined with a spallation source installed inside the subcritical core. The purpose of the TEF-P is roughly divided into three subjects:

- reactor physics aspects of the subcritical core driven by a spallation source;
- demonstration of the controllability of the subcritical core including a power control by the proton beam power adjustment;
- investigation of the transmutation performance of the subcritical core using certain amount of MA and LLFP.

These experimental items are more fully described in the following sections.

For the preparation of fuel, because a large amount of low-enriched uranium or plutonium is necessary to make the core critical or near-critical (*e.g.* k_{eff} = 0.95) in the fast neutron system, we expect to use the plate-type fuel of the FCA with various simulation materials such as lead and sodium for coolant, tungsten for solid target, ZrH for moderator, B_4C for absorber and AlN for simulating nitride fuel. Therefore, the TEF-P is designed with reference to FCA; the horizontal table-split type critical assembly with a rectangular lattice matrix. The proton beam can be introduced horizontally from the centre of the fixed half assembly.

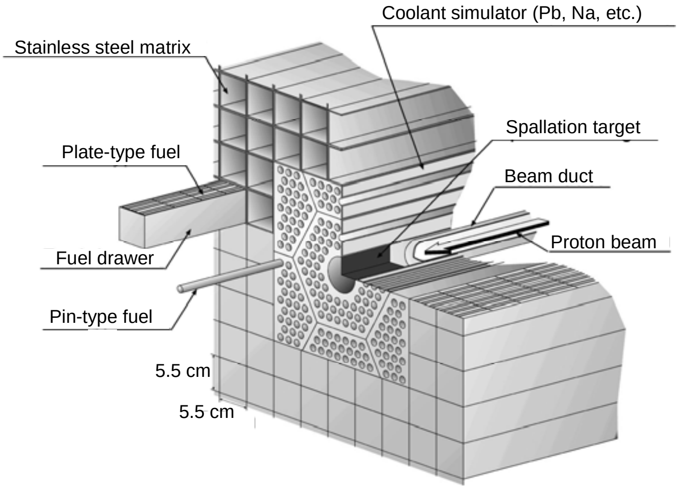
In the experiment with proton beam, the k_{eff} of the assembly will be kept less than 0.98. One proton with an energy of 600 MeV produces about 15 neutrons by the spallation reaction with heavy metal target such as lead. The 10 W proton beam corresponds to the source strength of 1.5×10^{12} neutrons/s, which have enough strength to measure the power distribution at the deep subcritical state such as $k_{eff} = 0.90$. The unexpected introduction of the 10 W proton beam into the critical state can be terminated safely with the reactor scram.

From the viewpoint of the neutronics of the subcritical system, subcriticality, power distribution, effective neutron source strength, and neutron energy spectrum will be measured by changing k_{eff} and spallation neutron source position, parametrically. The target material can be altered with Pb, Pb-Bi, W and so on. The reactivity worth is also measured for the case of coolant void and coolant infiltration into the beam duct. It is desirable to make the core critical in order to ensure the quality of experimental data of the subcriticality and the reactivity worth. For the demonstration of hybrid system, feedback control of the fission power is examined by adjusting the beam intensity. Operating procedures at start-up/shutdown, beam trip and re-start will also be examined.

As for the transmutation characteristics of MA and LLFP, fission chambers and activation foils are used to measure the transmutation rates. The cross-section data of MA and LLFP for the high-energy region (up to several hundred MeV) can be measured by the time-of-flight (TOF) technique with the proton beam of about 1 ns pulse width. Several kinds of MA and LLFP samples are also prepared to measure their reactivity worth, which is important for the integral validation of cross-section data.

The installation of a partial mock-up region of MA nitride fuel with air cooling is considered to undertake the measurement of the physics parameters of the transmutation system. The central rectangular region (28 cm × 28 cm × 60 cmL) will be replaced with a special subassembly which can install the pin-type MA fuel around the spallation target as shown in Figure 3.

Figure 3: TEF-P assembly with partial loading of pin-type fuel



Outline of TEF-T

In the current JAEA study, the Pb-Bi target/cooled ADS is a primary candidate. To solve technical difficulties for Pb-Bi application, TEF-T plans to complete the database required for Pb-Bi target/cooled ADS design. The experiments to obtain the material database for beam window are also the important mission of TEF-T.

TEF-T mainly consists of a Pb-Bi spallation target, a secondary cooling circuit and an access cell to handle the spent target vessel and irradiation test pieces. Pb-Bi is filled into a double annular cylindrical tube made by type 316 stainless steel. An effective size of the tube is about 15 cm diameter and 3 to 4 meters long, as shown in Figure 4. The target head is designed to be changeable according to the objective of the experiment. One of the target heads is designed to irradiate samples in the flowing Pb-Bi environment. A primary Pb-Bi loop is designed to allow Pb-Bi flow up to 2 meters per second of velocity and 450°C of the maximum temperature. Pb-Bi is circulated by an electromagnetic pump (EM pump) that is independent from the target tube. So, target unit can be replaced quickly and easily by withdrawing the target tube in an upward direction from the heat exchanger electromagnetic flow meter and EM pump.

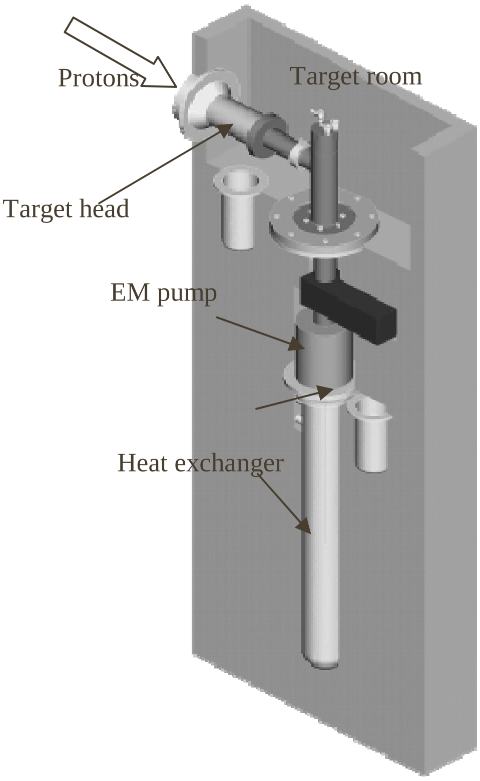
Activities for TEF-P construction

According to the current construction schedule, TEF will be built in two phases: TEF-P with a 30 kW class beam dump at first phase and TEF-T at latter phase. To design the facility, various R&D are now under way.

Low-power proton beam extraction

For the subcritical experiments using TEF-P, incident proton beam must be of the order of 10 W. It is also important to keep and represent the experimental condition of injected proton beam. To extract a very low-power proton beam constantly from the high-power J-PARC accelerator, development of the laser charge exchange technique is performed. By using laser beam, H^- beam from J-PARC linac changes into neutral and these neutral particles are easily separated from main stream H^- beam because the neutral beam does not sense the magnetic field of bending magnet. It is also studied to avoid the pre-neutralised beam generated from collision of H^- beam and remaining gas in accelerator tube to improve the quality of the low-power proton beam.

Figure 4: Sealed annular-type spallation target



A first demonstrative test of the laser charge exchange technique was performed using J-PARC Medium Energy Beam Transport [6]. The Nd:YAG laser using a pulse width of 20 nsec, beam energy of 500 mJ (repetition rate of 25 Hz) has been installed. At the experiment, the photo-stripped electron signal corresponded to the reduction of the total beam current signal at downstream were observed. The results of transverse profile measurements were also consistent with wire scanner signals of upper and downstream. The H^- beam components intercepted by a 0.8 mm height laser beam have been estimated by transverse profile measurement, and agree with photo detached fraction (Faraday cup and current detector's signal). The calculation results also show the complete (>99%) neutralisation ratio with 130 mJ Nd:YAG laser for 3 MeV H^- beam. Thus the complete (>95%) photo neutralisation fraction for a 130 mJ (repetition frequency of 5 Hz) 1 064 nm Nd:YAG laser pulse on a 15 mA, 3 MeV H^- beam could be confirmed practically.

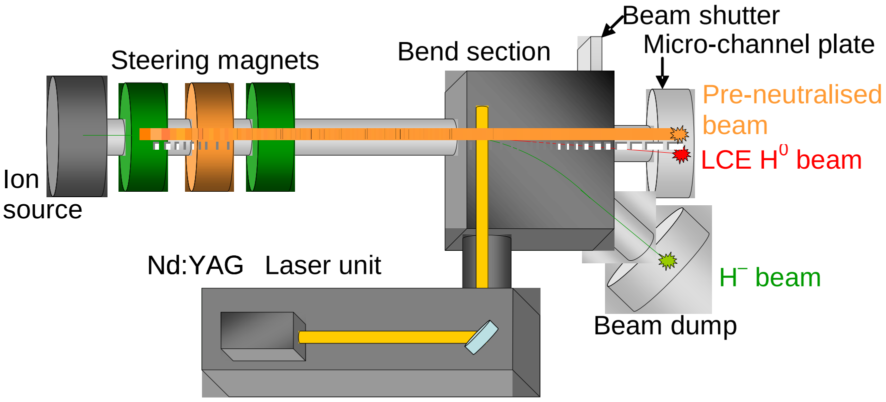
The second test was carried out at the JAEA-Tokai site. A prototype H^- ion source for J-PARC linac (70 keV-5 uA) and the same Nd:YAG laser source were used. A bending magnet with laser injection port was newly attached to the test line to extract neutralised beam during the beam bending. Figure 5 shows the experimental layout. There were limited beam control devices (two steering magnets), the beam was defocused and 50% of the proton beam was already neutralised at the exit of the bending magnet. However, by using a movable beam slit in front of the micro-channel plate detector to suppress the pre-neutralised beam, the neutralised proton beam signal related to the laser injection trigger was observed.

Minor actinide fuel handling method

To use MA-bearing fuel in TEF-P, the availability and limitation were studied. A trial manufacture of the coolant simulation block and conceptual study of remote fuel loading method of the pin-type MA fuel were performed. By using specified MA fuel loading zone configuration, effectiveness of the integral experiments using MA fuel were evaluated.

To prepare MA-bearing fuel, availability and experiences of MA usage in JAEA were surveyed and there were actual achievement by inclusion of Np and ^{241}Am into MOX fuel up to 5%. Irradiation of

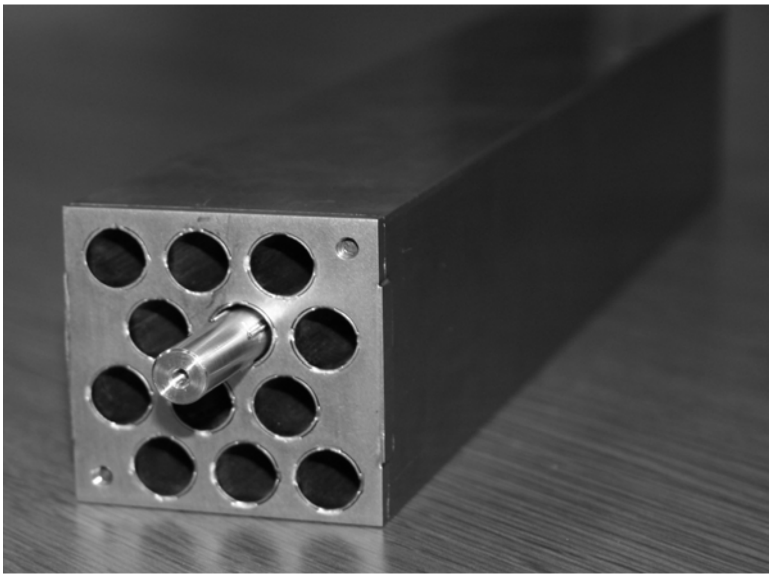
Figure 5: Layout of second laser charge exchange test



5% MA-bearing fuel in JOYO was performed and the soundness of the fuel pellet was proved. That means the fuel is of sufficient quality to use in zero-power critical assembly. From the viewpoint of actual experiences, preparation of low ^{237}Np and/or ^{241}Am contained (up to 5%) MOX fuel is reasonable as a first step of MA usage. To simulate neutron energy spectrum of ADS/FR with MA-bearing fuel, about 0.03 m^3 of driver zone (25 cm × 2 5cm × 60 cmL) is required. The effectiveness of integral experiments using MA-bearing fuel in the driver zone was quantified [7].

To load a pin-type fuel into FCA-type critical assembly, coolant simulation block is required which has a high coolant filling factor and appropriate air gap to cool MA pin fuel. Trial manufacture of coolant simulation block to simulate lead coolant was performed. Figure 6 shows the trial coolant simulator block. About 97% of coolant filling factor in simulation block was realised and can be increased by improvement of the manufacturing accuracy around the fuel pin guide tube.

Figure 6: Trial coolant simulation block



Even with a 5% addition of MA, a remote handling device to handle the MA-bearing fuel is required to protect the experimenters from radiation exposure. However, to maintain ideal experimental conditions, the handling head of the fuel pin must be manufactured as small as possible. A mechanical method using a small rubber was tested using the lead coolant simulation block mentioned above and enough performance to handle the pin-type fuel was demonstrated.

Establishment of user community

To clarify the needs to construct TEF, the Project Team called for preliminary Letters of Intent (LOI) for experiments at the TEF in the year of 2006. The purposes of preliminary LOI are:

- to know which groups have an interest in this activity and what contributions from them can be expected;
- to reflect new ideas and proposals on the specifications and the layout of TEF including the beam dump;
- to establish an appropriate collaboration scheme between J-PARC and the anticipated outside users.

As of August 2008, 38 proposals had been received in total. More than 10 experimental proposals were collected for both ADS and MA-loaded FBR, respectively. In other fields, six proposals for nuclear data measurements, three for high energy physics, two for LBE spallation target technologies, and various researches using protons and neutrons at the beam dump were proposed. Although the detailed discussions for the proposals have not yet begun, it is clear that TEF can serve as a basic experimental platform for nuclear science, engineering and applications. The project is still open to accept other proposals. The participants who are interested in experiments at TEF-P are many and varied. From overseas, EUROTRANS, Switzerland, China, Korea, Serbia and Malaysia provided letters of interest for TEF. Nine Japanese universities and one commercial company also presented proposals. We are still open to receiving proposals.

To encourage the activities to construct TEF, a research committee on experimental facility for reactor physics concerning actinide management was founded in August 2008 by the Atomic Energy Society of Japan. The objectives of the committee are:

- to survey the needs of neutronic experiments necessary for actinide management;
- to specify the requirement not only for experimental methods, devices and equipments but also experimental facility design;
- to clarify issues for effective use of the experimental facility in the field of scientific research, education and publicity.

The discussion of the committee will be summarised as recommendations for the newly planned facility, TEF-P. More than 30 reactor physics experts from universities, research institutes and heavy industrial companies were present. It is scheduled that the recommendation to design the TEF-P facility will be proposed within two years.

Conclusion

To perform the design study of the transmutation system for long-lived nuclides, the construction of TEF, which consists of two buildings, TEF-P and TEF-T, is proposed under the J-PARC Project.

TEF-P is a critical assembly, which can accept the 400 to 600 MeV-10 W proton beam for the spallation neutron source. The purposes of TEF-P are the experimental validation of the data and method to predict neutronics of the fast subcritical system with spallation neutron source, the demonstration of the controllability of subcritical system driven by an accelerator, and the basic research of reactor physics for transmutation of MA and LLFP. Distinguishing characteristics of the TEF-P in comparison with existing experimental facilities can be summarised as follows: *i)* both the high-energy proton beam and the nuclear fuel are available; *ii)* the maximum neutron source intensity of about 10^{12} n/s is strong enough to perform precise measurements even in the deep subcritical state (*e.g.* $k_{eff} = 0.90$), and is low enough to easily access the assembly after the irradiation; *iii)* a wide range of pulse widths (1 ns-0.5 ms) can be made available through the laser charge exchange technique described earlier; *iv)* MA and LLFP can be used as a shape of foil, sample and fuel by installing an appropriate shielding and remote handling devices.

TEF-T is a facility to prepare the database for engineering design of ADS using a 600 MeV-200 kW proton beam and the Pb-Bi spallation target. The purposes of TEF-T are R&D for the irradiation damage of the beam window, the compatibility of the structural material with flowing liquid Pb-Bi and the operation of the high-power spallation target.

Along with the design study of the TEF, R&D for the components required for TEF, such as laser charge exchange technique to extract very low-power proton beam and test manufacturing of MA fuel handling devices were performed. From the experimental result of the laser charge exchange technique, beam extraction in the magnetic field is successfully demonstrated with J-PARC prototype ion source. The significant improvement of analysis accuracy of actual ADS was expected by critical experiment with MA fuel at TEF-P.

Preliminary letters of intent were called for to encourage the project. Over a hundred research projects have been proposed for the experimental programme using TEF. A research committee on an experimental facility for reactor physics concerning actinide management was founded by the Atomic Energy Society of Japan, and recommendations reflecting the Japanese reactor physics researchers' community will be presented after two years of discussion. The results of these activities will be reflected in the facility design of TEF.

Acknowledgements

An important experiment of laser charge exchange technique in magnetic field was done by the great effort of the members of J-PARC Center, K. Hasegawa, N. Ouchi, T. Tomizawa and F. Hiroki. N. Aoki and N. Kobayashi of Toshiba Corporation also made the efforts for design and preparation of laser charge experiment devices. The authors are grateful for the support of these persons and companies.

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Reactivity monitoring of the YALINA subcritical assembly using beam trips and current-to-flux experiments

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Abstract

Transmutation of spent nuclear fuel is a key technology for sustainable nuclear energy, the ADS being one of the concepts explored to reduce its radiotoxicity and volume. In the design of future ADS facilities, the reactivity monitoring system is of highest importance. Hence, it is necessary to determine the best reactivity determination techniques and the required electronic chains for the peculiarities inherent to ADS. Within the international project IP-EUROTRANS a set of experiments are being carried out in the subcritical facility YALINA-Booster. This paper presents the results from an experimental campaign including pulsed neutron source, current-to-flux and beam trip measurements based on both pulsed and current mode electronic chains.

Introduction

In the framework of IP-EUROTRANS [1], within the 6th FW Programme of the UE, an experimental campaign has begun at the YALINA-Booster facility located at JIPNR-Sosny (Belarus). The YALINA experiments will continue the work performed during the MUSE programme (UE 5th FW Programme). The main objective is the qualification of the reactivity monitoring techniques as well as to develop the electronic chains that can be used in a power ADS. For these purposes, YALINA couples a D-T neutron generator to a flexible zero-power subcritical assembly with an intermediate neutron spectrum. The high intensity of the accelerator and the possibility to work in continuous or pulsed modes allow the study of the current-to-flux relationship, beam trip experiments and dedicated experiments for loading and start-up procedures. In addition, it will provide the opportunity to test the electronic chains in current mode, corresponding to the most probable condition in a power ADS. Reactivity determination with pulsed mode techniques has been tested widely in several subcritical experiments [2-4], but current mode, being the most suitable measurement technique at high power, has, to the knowledge of the authors, not been tested in terms of for instance the area ratio and beam trip techniques.

Experimental set-up

The YALINA-Booster is a subcritical fast-thermal core coupled to a neutron generator (NG-12-1). The neutron generator uses a deuteron ion accelerator impinging on a Ti-T or Ti-D target to produce fusion neutrons. With a diameter of 45 mm, the target is located in the centre of the core. In the experiments presented here only the Ti-T target was used, thus providing a quasi-isotropic neutron energy spectrum of 14 MeV. The neutron generator can be operated in both continuous and pulse modes and gives thereby the possibility of performing both pulsed neutron source (PNS) measurements and continuous wave measurements. Moreover, the continuous wave can be promptly interrupted (~1 μs) followed by a fast beam restart. In this way, short repeated beam trips can be induced intentionally with interruption times in the millisecond scale. The maximum beam current in continuous mode is around 1.5 mA, giving a maximum neutron yield of approximately 10^{11} neutrons per second.

The core [5], depicted in Figure 1, consists of a central lead zone (booster), a polyethylene zone, a radial graphite reflector and a front and back biological shielding of borated polyethylene. The booster zone has been loaded with two different arrangements of 36% enriched UO_2 pins while the polyethylene is loaded with 10% enriched UO_2 fuel pins. The SC3a configuration presented here is based on 1 077 10% enriched fuel pins and has a k_{eff} ~0.94.

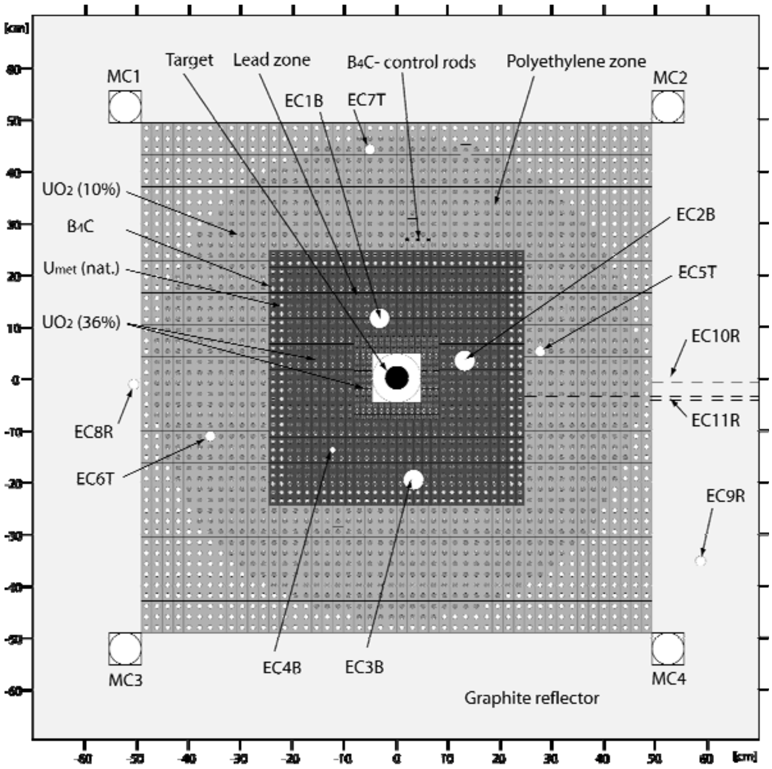
The fast spectrum lead zone and the thermal spectrum polyethylene zone are separated by a so-called thermal neutron filter, or valve zone, consisting of one layer of 108 pins with metallic natural uranium and one layer of 116 pins with boron carbide (B_4C), which are located in the outermost two rows of the fast zone. Hence, thermal neutrons diffusing from the thermal zone to the fast zone will either be absorbed by the boron or by the natural uranium. In this way, a coupling of mainly fast neutrons between the two zones is maintained.

The three B_4C-control rods that can be inserted in the thermal zone have allowed us to slightly change (-0.5$) the reactivity of the system. Hence, the sensitivity of the different reactivity monitoring techniques can be tested.

There are seven axial experimental channels (EC1B-EC4B and EC5T-EC7T) in the core, two axial (EC8R and EC9R) and two radial experimental channels (EC10R and EC11R) in the reflector. In addition, there is one neutron flux monitoring channel in each corner of the core (MC1-4) and outside the reactor (Y.T. not shown in the figure).

Two types of detectors can be placed in these experimental channels to monitor the flux, ^{235}U fission chambers and ^{3}He detectors. Both pulsed mode electronic chains and current mode electronic chains have been used throughout the experiments. The main purpose has been to verify that the proposed reactivity monitoring techniques may also be used in current mode detection. A crucial point when measuring in current mode is to achieve high enough signal to noise ratio. When running with the Ti-T target at the highest current, it was possible to achieve a count rate in the corresponding pulsed electronic chain at about 10^6 counts per second when using a 500 mg fission chamber in the booster zone. This count rate was high enough to give a signal in the current mode electronic chain after reducing the background noise level.

Figure 1: Schematic cross-sectional view of the YALINA reactor core for the SC3a configuration



PNS experiments

In the PNS experiments, the kinetic evolution of the system is measured after the repetitive injection of a neutron pulse. Two methods have been used to obtain the reactivity from the PNS measurements: the area method [6] (also referred to as the Sjöstrand method) and the determination of the prompt decay constant [7].

Figure 2 shows the kinetic evolution of the neutron flux measured at different locations of the reactor to a neutron pulse of 5 μs with a repetition rate of 50 Hz. Inspecting the figure we can observe the different evolutions obtained at the booster (EC1B) or the innermost thermal zone (EC5T), which present a fast decay during the first millisecond followed by a slow decay, compared with the evolutions at the graphite reflector (MC2) or outside the reactor (Y.T.), which increase the level over the first two milliseconds to decay slowly afterwards. It should be pointed out that in order to reduce statistical uncertainties it has been necessary to accumulate the kinetic response from a large number of pulses. In addition, due to the high counting rates observed in some of the detectors, it was necessary to take into account the effect of the dead time, leading to corrections in the peak amplitude up to 25% for the highest intensities.

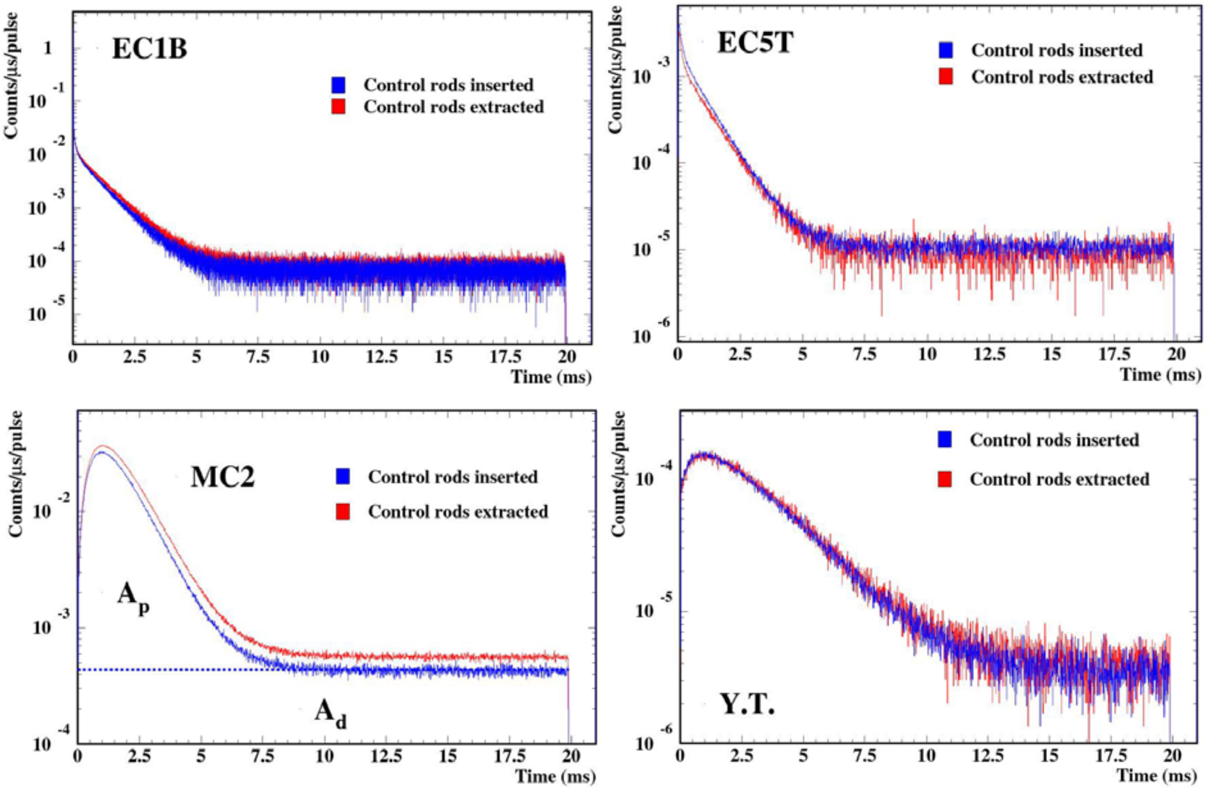
Prompt decay constant method

From the point kinetic approximation, we find that the kinetic evolution of prompt neutrons after a pulse follows an exponential decay with decay constant, α:

$$\alpha = \frac{\rho - \beta_{eff}}{\Lambda} \Rightarrow \frac{\rho}{\beta_{eff}} = \frac{\alpha}{\beta_{eff}/\Lambda} + 1$$

where β is the reactivity, β_{eff} the effective delayed neutrons fraction and Λ the mean neutron generation time. Hence, if the value β_{eff}/Λ is known, the value of ρ (in units of β_{eff}) can be calculated.

Figure 2: Kinetic evolution of the neutron flux measured at different locations in YALINA SC3a configuration to a 5 μs pulse and 50 Hz repetition rate. The plots show accumulated statistics for about 10^5 neutron pulses.



The time response to 14 MeV neutrons for all the detectors has been simulated with MCNP [8]. With these responses, it was possible to correct the reactivity values obtained with the slope fit method. It was found that the correction decreases the reactivity values in 0.9 \$, approximately. The value of $\beta_{eff}/\Lambda = 114 \pm 2$ s^{-1} has been also obtained from MCNP calculations. With this constant, the values of the reactivity obtained with the slope fit method are shown in Table 1.

Table 1: Reactivity values[1] obtained from the prompt decay constant for different regions of the reactor. The measurements were performed with the control rods inserted and extracted.

		Control rods extracted		Control rods inserted	
		α (s^{-1})	ρ (\$)	α (s^{-1})	ρ (\$)
Booster zone	EC1B	-1026 ± 17	-7.97 ± 0.19	-1096 ± 21	-8.58 ± 0.22
	EC2B	–	–	-1087 ± 16	-8.50 ± 0.19
	EC3B	–	–	-1090 ± 4	-8.53 ± 0.13
Thermal zone	EC5T	-1031 ± 26	-8.01 ± 0.26	-1107 ± 37	-8.68 ± 0.34
	EC6T	–	–	-1089 ± 15	-8.52 ± 0.18
Graphite reflector	MC2	-976 ± 6	-7.53 ± 0.12	-1018 ± 8	-7.89 ± 0.14
Corrected average			**8.7 ± 0.3**		**9.3 ± 0.3**

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[1] In this work the quoted errors include only statistical uncertainties from both data and/or Monte Carlo simulations, whereas systematic effects are not included.

Area method

The area method states that the reactivity, in dollars, can be determined as follows:

$$\frac{\rho}{\beta} = -\frac{A_p}{A_d} = 1 - \frac{A_t}{A_d}$$

where A_p is the area under the pulse that is due only to the prompt neutrons generated after the pulse, A_d the area due only to the delayed neutrons and A_t is the total area under the curve, as shown in the bottom left panel of Figure 2.

Table 2 shows the reactivity values obtained with the area method for detectors located in different positions within the experimental set-up. Inspecting the table, large differences can be observed among detectors located in the booster and the thermal or reflector zones. These differences arise from the different positions relative to the neutron source and the different neutron absorption cross-section in the different reactor regions and close to the detectors. Nevertheless we observe that the reactivity reaches a stable value when the distance from the detector to the neutron source is large enough.

Table 2: Reactivity values directly obtained with the area method from different detectors. The measurements were performed with the control rods inserted and extracted.

		Control rods extracted	Control rods inserted
Booster zone	EC1B	-14.93 ± 0.17 $	-17.17 ± 0.22 $
	EC2B	–	-15.30 ± 0.15 $
	EC3B	–	-10.18 ± 0.04 $
Thermal zone	EC5T	-8.77 ± 0.35 $	-9.46 ± 0.21 $
	EC6T	–	-7.57 ± 0.14 $
Graphite reflector	MC2	-7.26 ± 0.03 $	-7.88 ± 0.06 $
	MC3	-7.33 ± 0.96 $	-7.96 ± 1.13 $
Outside reactor	Y.T.	-7.41 ± 0.23 $	-8.01 ± 0.23 $

To estimate the reactivity of the system we compare these values with the results obtained from MCNP simulations. The reactor has been simulated using a detailed description of the SC3a geometry and JEFF-3.1 libraries. Two types of calculations were performed: First, a criticality calculation providing $k_{eff} = 0.94906 \pm 0.00009$, $\beta_{eff} = 683 \pm 9$ pcm and $\Lambda = 59.7$ μs, which lead to a reactivity of $\rho_{MCNP} = 7.86 \pm 0.10$ $. Second, a source calculation to obtain the estimated counting rates at the detector locations. This calculation was performed including (or not) delayed neutrons, respectively, allowing to simulate the area method with the MCNP code. Normalising the simulated area ratio with the value of ρ_{MCNP} obtained from the simulations, we obtained the correction factors for each detector, shown in Table 3.

Table 3 also shows the corrected values of the reactivity. As can be observed, all the values become compatible after the corrections, leading to a reactivity value of ρ_{CRE} ($) =-9.0 ± 0.2 and ρ_{CRI} ($) = 9.7 ± 0.2. It is important to note that, although both values of the reactivity are compatible due to the uncertainties, the correction process is introducing a large correlation between the reactivity values, which reduces the uncertainty in the difference.

Table 3: Correction factors, calculated with MCNP, for the reactivity values at different locations of the reactor core calculated with the area method.

Experimental channel	Correction factor	ρ ($), CRE	ρ ($), CRI
EC1B	1.71 ± 0.06	-8.7 ± 0.3	-10.1 ± 0.4
EC2B	1.54 ± 0.05	–	-9.9 ± 0.4
EC3B	1.06 ± 0.04	–	-9.6 ± 0.3
EC5T	0.87 ± 0.04	-10.1 ± 0.6	-10.9 ± 0.6
EC6T	0.87 ± 0.04	–	-8.7 ± 0.4
MC2	0.81 ± 0.03	-9.0 ± 0.3	-9.8 ± 0.4
MC3	0.86 ± 0.03	-8.5 ± 1.2	-9.3 ± 1.4
Average		**-9.0 ± 0.2**	**-9.7 ± 0.2**

Current-to-power measurements

A key point of the future ADS facilities will consist of a robust on-line and continuous monitoring of the subcritical assembly reactivity. This monitoring system must yield valuable information concerning the rapid relative change of the reactivity, which in a subcritical assembly is given by:

$$-\rho = q * \frac{S}{P}$$

where q is the energy released per neutron fission, S is the source strength, P is the thermal power (neutron flux) and φ^* is the source importance. It is generally assumed that the source strength is proportional to the beam current and, hence, any change in the reactivity is accessible through the beam current ($S = \kappa \cdot I$) and the neutron flux (P). The on-line determination of the reactivity, then, requires the monitoring of three quantities, the accelerator intensity (I), the neutron source intensity (S) and the core power (neutron flux) P. Actually two ratios must be determined, S/I and P/S. However, there exist several factors which can affect this proportionality, such as the impinging position of the beam at the tritium target, the beam emittance, the beam oscillations or even the target consumption.

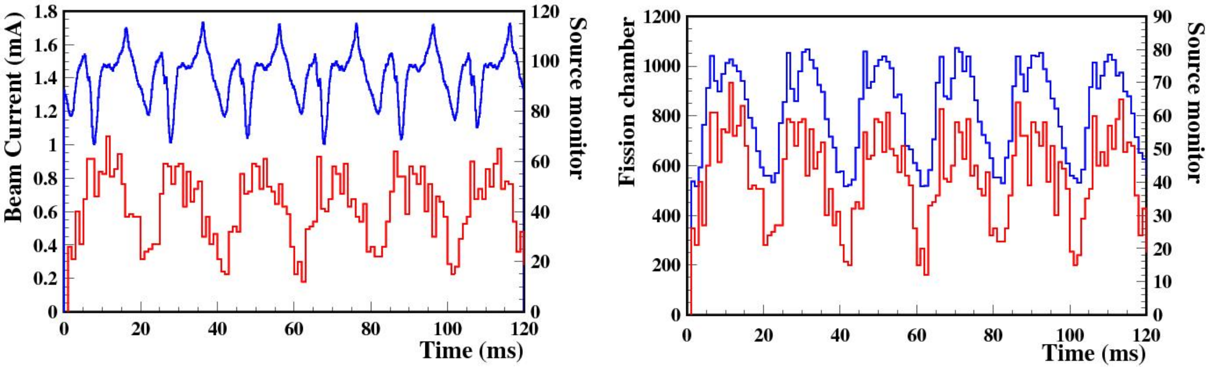
In the YALINA experiments, the determination of the neutron source intensity relies in measuring neutrons coming directly from the D-T fusion reactions (14 MeV), which have energies larger than the fission neutrons created within the reactor. In this work we have used a BC501A [9] liquid organic scintillator acting as a veto detector to measure high-energy neutrons coming from the D-T source. The threshold detection energy was fixed by using a typical neutron fission spectrum from a ^{252}Cf source. In this sense, using a constant fraction discriminator (CFD) we were able to neglect any neutron with energy below ~7 MeV.

The proportionality between I, S and P was already determined in the YALINA-booster during the experimental campaign of summer 2007. The data from the present measurements, once completely analysed, should test this proportionality over a wider source intensity range.

Within the EUROTRANS measurements at YALINA, we will and have explored several factors that can spoil the proportionality between I, S and P. In Figure 3 we show an example where we have lost the proportionality S/I while we keep P/S. In this experiment we observe a beam trip where we have monitored the beam current, the neutron source and a fission chamber. In the left panel, it is easy to appreciate that although the monitoring of the current and the monitoring of the neutron source show a 50 Hz oscillation, they differ in shape and magnitude. Actually, it is when we compare the neutron source monitor with the fission chamber (right panel) that we realise that the beam current is not proportional to the neutron source production whereas the neutron source monitoring system seems to work properly. It is worth stressing that this effect has been detected thanks to the high efficiency of the neutron source monitor, which allows us to measure the source every millisecond.

In addition, we have also recorded events were the beam current is proportional to the core power, but this proportionality is lost in the neutron source monitoring.

Figure 3: (Left) Time evolution of the deuteron beam intensity (blue line) and the neutron source monitoring system (red line). (Right) Time evolution of the fission chamber counting rate (blue line) and the neutron source monitoring system (red line).



Beam trip measurements

Current-to-flux technique can only provide relative measurements of the reactivity. Therefore, it is necessary to use absolute reactivity determination techniques to calibrate the former technique. To limit the perturbation to the power operation of ADS, we can take profit from the unavoidable occurrence of beam trips.

One of the most powerful advantages of beam trips to calibrate the reactivity while in operation is the possibility to apply source jerk [10] or prompt decay constant determination in a single beam trip. However, it is necessary to provide a neutron detection system capable of recording enough information during the beam trip. Usual pulsed mode electronic chains can limit the accuracy of the measurement due to the effect of dead time when we want to increase statistics. Hence, during the YALINA experiments, a large effort has been made at CIEMAT to prepare an electronic chain based on current mode detection for measuring the beam trips. The main characteristic of this chain is the possibility to reach a sampling rate as large as 125 MHz with 14-bit resolution. Nevertheless, in order to compare the results obtained with both chains we have kept several detectors working in pulsed mode detection.

The source jerk technique is based on the determination of the kinetic parameters after removal of the prompt neutron source, in analogy to the rod-drop technique. The main advantage of this technique is related to the fact that no fitting, based on an interpretation model, must be performed. On the other hand, corrections may be needed to take into account energy and spatial effects. However, since this technique is equivalent to the PNS area method, it is enough to obtain the correction factors for the latter.

The main advantage of determining the reactivity through the prompt decay constant is that the investigated period is much shorter than in the case of a source jerk since only the die-away of the prompt neutron population must be recorded. The drawback is that in the case of fast reactors with large reflectors, which is not the case of YALINA, but was that for instance of the MASURCA reactor during the MUSE-4 experiments, the decay of the prompt neutron population cannot adequately be represented by a single exponential. Therefore more complex interpretation models are needed and simulations may be required.

Figure 4 shows a typical beam trip measured in YALINA with the control rods extracted. In the left panel, we can observe the beam trip measured with a current mode detector located in the graphite zone of YALINA-Booster. As in the case of current-to-flux measurements (see Figure 3), we observe the large fluctuation produced by the oscillation of the neutron source. It is important to stress that due to the relatively low intensity of the source, the current in the 500 mg ^{235}U fission chamber is 1 μA at maximum, which obliged the use of amplification factors of 10^5-10^6 V/A. In fact, the original signal was measured at a 10 MHz sample rate, but it has been filtered with a 100 kHz low-pass filter to reduce electronic noise introduced in the signal due to the large amplification. This situation is unlikely to happen at high power, when the current in the detector is usually much larger and the amplification of pick-up noise should become negligible.

In addition to the current mode detector, we show, in the right panel of Figure 4, the same beam trip measured with a detector in pulsed mode. Inspecting the figure, one observes that the source jerk technique can be applied to this detector. However, the determination of the prompt decay constant cannot be achieved. In addition, one must take into account that, due to the high counting rate, the applied dead time corrections were large (~50% at maximum), and will increase the systematic uncertainties of the result.

Despite the low current and the oscillations, if the average over several oscillation cycles is performed prior to the beam trip one can obtain a fairly good estimation of the reactivity using the source jerk technique. The results using both current and pulsed modes provide compatible results with those obtained using the area method (see Table 2). The main argument to perform this type of averaging is that the delayed neutron fraction does not change during the oscillation and then, it will be proportional to the mean value of the prompt neutron level.

In order to reduce statistical uncertainties we have averaged several beam trips, as presented in Figure 5. This procedure has also allowed us to obtain a good estimation of the prompt decay constant. Inspecting the figure, we can see (in the left panel) the results of a beam trip with the insertion or extraction of the control rods. Comparing these results with the results of the area method, we can observe that the source jerk technique provides similar values.

Figure 4: Beam trip sequence measured with a fission chamber operating in current mode at the graphite zone (left) and in pulse mode at the booster (right panel), both with the control rods extracted

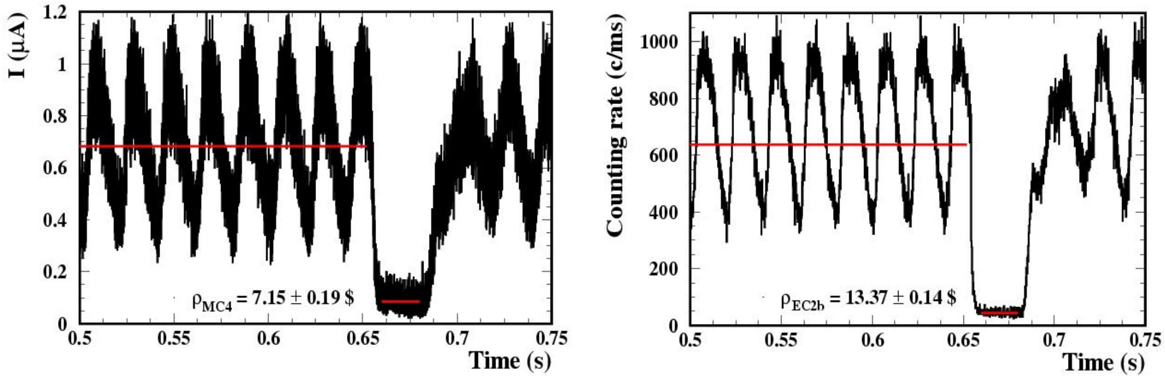
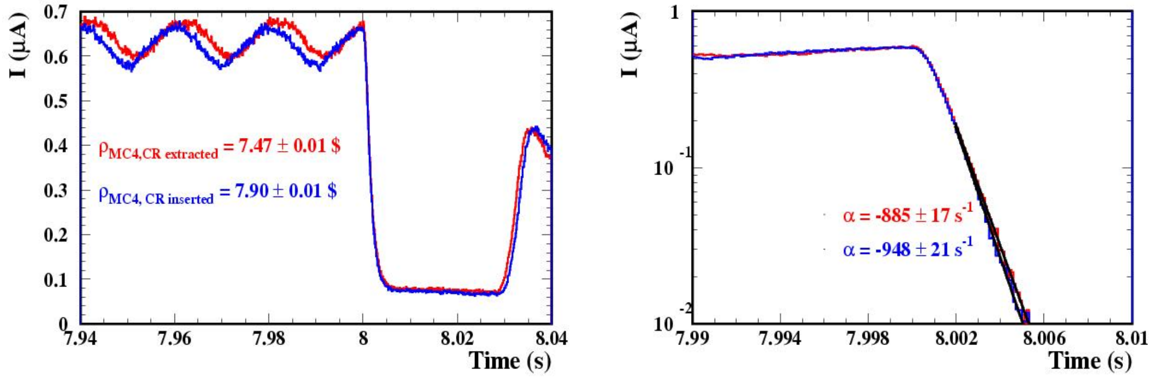


Figure 5: Averaged beam trip sequence with control rods inserted and extracted. (Left) Source jerk technique. (Right) Delayed neutron level has been subtracted to appreciate the different decay constant.



On the other hand, if we observe the right panel we can appreciate the prompt decay after subtracting the delayed neutron level. It is possible to see that the decay is different when we insert or extract the control rods, leading us to detect small variations of the reactivity. However, it is important to note that, at the present stage of the analysis, the observed prompt decay constant is slightly lower than in the PNS experiments. We are still investigating this effect.

Conclusions

In this work we have presented a preview of the experimental results obtained during the first phases of EUROTRANS experiments at the YALINA-Booster facility. The main objective has been to demonstrate the possibility of monitoring the reactivity of the system using short beam trips.

Using PNS techniques, we have demonstrated that the area and prompt decay constant fit methods for reactivity determination provide compatible results, after applying the necessary corrections computed with MCNP.

The 14 MeV neutron source was monitored with a liquid scintillator at intervals as short as 1 ms, showing that the intensity of the neutron source is not always proportional to the beam current of the deuterium accelerator.

We were working on a high-noise environment that affects especially the analogue measurements, like the beam current and the fission rate from the detectors operated in current mode. Despite the adverse experimental conditions we were able to make useful recordings of these fission chambers working in current mode, with current levels below 1 μA. Using the source jerk techniques we were able to estimate the reactivity of the system in current and pulse modes, from one or few short beam trips. To our knowledge, it is the first time that detectors in current mode have been used to determine the reactivity values in a subcritical assembly. The results obtained with this technique were compatible with those obtained by standard pulsed detectors, demonstrating the capabilities of the beam trip techniques to calibrate the reactivity for current-to-power measurements.

Acknowledgements

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Session VI

Transmutation systems: Design, performance and safety

Chairs: I-S. Hwang, K. McCarthy

Trends in transmutation performance and safety parameters versus TRU conversion ratio of sodium-cooled fast reactors

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Abstract

Transmutation performance and safety characteristics of sodium-cooled fast reactors were investigated for a wide range of transuranic (TRU) conversion ratios. Using ternary metal and mixed-oxide fuels, advanced burner reactor (ABR) core concepts of 1 000 MWt power rating were developed for target TRU conversion ratios of 1.0, 0.75, 0.50, 0.25 and 0.0. Two make-up TRU feeds were considered with minor actinides to plutonium ratios of 0.1 and 1.0. For all twenty design concepts with different TRU conversion ratio, fuel type and TRU feed, detailed fuel cycle analysis was performed and TRU consumption rate and isotopic mass flow rates were evaluated. Kinetics parameters and reactivity feedback coefficients were also calculated for the equilibrium cycle core configurations at the beginning and the end of cycle, and the integral reactivity parameters for the quasi-static reactivity balance analysis of the passive regulation of power were estimated. The results show that the TRU consumption rate reaches ~80% of the maximum theoretical value when the TRU conversion ratio is in the range of 0.25-0.35. As the TRU conversion ratio decreases, the initial heavy metal inventory and charge rate decrease significantly. On the other hand, the TRU inventory and charge rate increases, but to a lesser extent. The estimated safety parameters indicate that the metal and oxide cores of TRU conversion ratio in the range of 0.25-0.40 are feasible with favourable passive safety features.

Introduction

The advanced burner reactor (ABR) is one of the major technologies to be developed in the Global Nuclear Energy Partnership (GNEP). The primary mission of ABR is to demonstrate the transmutation of transuranics (TRU) recovered from light water reactor (LWR) spent fuel, and hence the benefits of the fuel cycle closure to nuclear waste management [1,2]. To achieve a high TRU consumption rate, it is desirable to have a low TRU conversion ratio. Here, the TRU conversion ratio is defined as the ratio of the net number of TRU nuclides produced over a cycle to the net number of TRU nuclides destroyed over a cycle. A low conversion ratio however requires a high TRU fraction in heavy metal, which is far beyond the current irradiation experience with plutonium-based fast reactor fuels. Many technical challenges have also been identified that must be overcome to utilise high TRU concentration fuel. In order to generate representative data useful in selecting an optimum TRU conversion ratio for different technical options, therefore, a systematic study was performed on the trends in transmutation performance and safety parameters versus TRU conversion ratio for sodium-cooled recycle reactors.

A range of ABR core concepts of 1 000 MWt power rating were previously developed [3], starting from the SuperPRISM (S-PRISM) core designs [4]. Both metal and oxide fuel cores were developed for TRU conversion ratios of 1.0, 0.75, 0.50, 0.25 and 0.0 using the TRU from LWR spent fuel. To allow for transition to higher or lower conversion ratios as required, the overall assembly dimensions were retained, while the number and size of fuel pins within each assembly were adjusted to achieve the target conversion ratio. Compact core designs were developed by removing the blankets of the S-PRISM design and by further reducing the core diameter in order to maximise the average linear power within the thermal design limits. It was also tried to maximise the discharge burn-up within the fast fluence limit of assumed cladding material.

By extending this ABR core design study to different TRU feeds with additional design modifications, the impacts of the TRU conversion ratio on the core performance and safety characteristics were investigated. Two different minor actinide (MA) to plutonium (Pu) ratios were considered: MA/Pu $\cong$ 0.1 as in the typical TRU recovered from LWR spent fuel and MA/Pu $\cong$ 1.0 as in the TRU from multi-recycled Pu fuel. For all twenty design concepts with different TRU conversion ratio, fuel type and TRU feed, detailed fuel cycle analysis was performed and TRU consumption rate and isotopic mass flow rates were evaluated. Kinetics parameters and reactivity feedback coefficients were also calculated for the equilibrium cycle core configurations at the beginning and the end of cycle, and the integral reactivity parameters for the quasi-static reactivity balance analysis of the passive regulation of power were estimated.

Development of metal and oxide core concepts

Starting from the S-PRISM core designs, metal and oxide core concepts of 1000 MWt ABR were developed for TRU conversion ratio (CR) from 0.0 to 1.0. Since TRU transmutation fuels are not fully developed, conventional design approaches were adopted to stay within current fast reactor technology bases as much as possible. A ternary metal fuel form of U-TRU-Zr and a mixed-oxide form of $(U\text{-}TRU)O_2$ were assumed for the metal and oxide cores, respectively. Low-swelling ferritic stainless steel (HT9) cladding was selected as structural material (cladding and duct). Key thermal-hydraulic and materials-related design constraints for fuel and structural material were imposed on selection of design parameters. The fuel smeared density was assumed 75% for the ternary metal fuel and 85% theoretical density for the oxide fuel. To stay within the ferritic cladding database, a cladding fast fluence limit was assumed to be about 4×10^{23} n/cm^2.

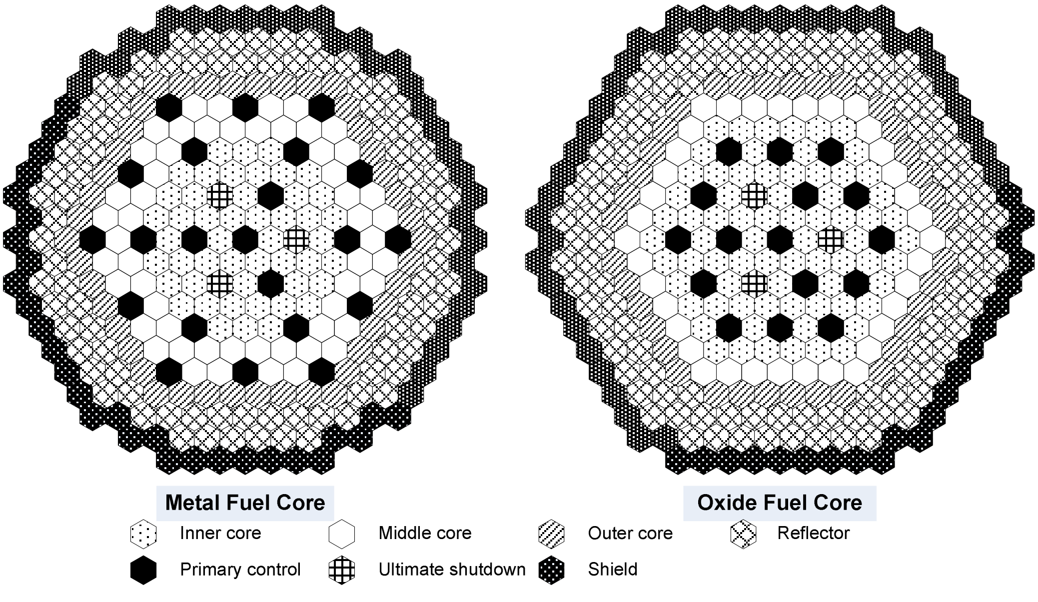
Both metal and oxide core designs were developed for target TRU conversion ratios of 1.0, 0.75, 0.50, 0.25 and 0.0 at an equilibrium cycle in which the TRU recovered from ABR spent fuel was used as the primary feed and the TRU from an external supply was used for make-up. Two make-up TRU feeds were considered with different minor actinides to plutonium ratio. One is the TRU recovered from five-year cooled LWR spent fuel with average discharge burn-up of 50 MWd/kg (MA/Pu $\cong$ 0.1) and the other is the TRU from multi-recycled Pu fuel (MA/Pu $\cong$ 1.0). The isotopic compositions of the make-up TRU feeds are shown in Table 1. The make-up uranium was assumed to be depleted uranium. For the uranium-free fuel for zero TRU conversion ratio, no make-up uranium was used, but it was assumed that all the *in situ* produced uranium, mostly ^{234}U, is recycled. As a result, the calculated conversion ratio was slightly above zero at equilibrium and the TRU fraction was slightly less than 100%.

Table 1: Isotopic composition (wt.%) of make-up TRU feeds

TRU from LWR spent fuel				TRU from multi-recycled Pu fuel			
^{237}Np	4.8	^{241}Am	3.4	^{237}Np	7.3	^{241}Am	18.8
^{236}Pu	0.0	^{242}Am	0.0	^{236}Pu	0.0	^{242}Am	0.1
^{238}Pu	2.3	^{243}Am	1.5	^{238}Pu	2.0	^{243}Am	15.9
^{239}Pu	47.9	^{242}Cm	0.0	^{239}Pu	18.2	^{242}Cm	0.0
^{240}Pu	22.5	^{243}Cm	0.0	^{240}Pu	13.4	^{243}Cm	0.1
^{241}Pu	10.6	^{244}Cm	0.5	^{241}Pu	5.9	^{244}Cm	7.0
^{242}Pu	6.5	^{245}Cm	0.0	^{242}Pu	10.6	^{245}Cm	0.9

Each of these ABR designs is a compact core developed to maximise the linear power within the thermal design limits. By removing the blankets of the S-PRISM design, the number of fuel assemblies (driver and blanket) was reduced by over 35% for the metal fuel core and by nearly 50% for the oxide fuel core. The core diameter was further reduced to increase the linear power within the thermal design limits. The basic ABR configuration consists of 144 fuel assemblies except for the break-even (CR = 1.0) cores that have 151 fuel assemblies. To compensate for the increasing burn-up reactivity swing, the number of primary control assemblies was increased up to 22 for low conversion ratio cores. Example metal and oxide core layouts are shown in Figure 1 for the TRU conversion ratio of 0.25. The fuel cycle length was also adjusted such that the burn-up reactivity swing is within the reactivity control capability of the primary control system.

Figure 1: Metal and oxide core layouts for a TRU conversion ratio of 0.25



For each target TRU conversion ratio, the fuel volume fraction was iteratively determined to yield the target conversion ratio that is primarily determined by the TRU enrichment, *i.e.* the TRU fraction in heavy metal (HM). The fuel pin size and the number of fuel pins per assembly were adjusted to yield the appropriate fuel volume fraction and satisfy the linear power limit. When the TRU fraction is larger than 30%, the Zr weight fraction in the U-TRU-Zr metal fuel was increased up to 40% in proportion to TRU fraction to compensate for the deteriorated thermal properties. The overall assembly dimensions were retained to allow for a transition to higher or lower conversion ratios as required; the assembly pitch was 16.14 cm, and the active core height was 101.6 cm for the metal cores and 137.2 cm for the oxide cores. Wire-wrap spacers were generally used, but for small diameter pins, grid spacers were assumed with seven tie pins. The same fuel assembly design parameters were used for both the TRU feeds, except for the uranium-free fuel cores (CR = 0.0).

For given fuel volume fraction and cycle length, the TRU enrichment was determined from equilibrium cycle analysis such that the multiplication factor at the end of cycle (EOC) is 1.0 with the control rods fully withdrawn from the active core. To flatten the radial power distribution, enrichment zoning strategy was employed except for the uranium-free fuel cores (CR = 0.0), where different fuel pin diameters were used. The number of fuel batches was adjusted to maximise the fuel residence time such that the discharge burn-up is maximised within the fast fluence limit of cladding. Tables 2 and 3 present the key core and fuel assembly design parameters of metal and oxide fuel cores.

Table 2: Key core and fuel assembly design parameters of metal fuel cores

Target TRU CR		**1.0**	**0.75**	**0.5**	**0.25**	**0.0**	
						MA/Pu ≅ 0.1	**MA/Pu ≅ 1.0**
Fuel assemblies		19/66/66[a]	30/42/72	42/66/36	48/54/42	78/24/42	78/24/42
Control assemblies		9/3[b]	16/3	16/3	22/3	22/3	22/3
Fuel pins per assembly		271	271	324	540	540/540/547	540/540/547
Fuel pin diameter (cm)		0.81	0.76	0.62	0.46	0.45/0.49/0.54	0.49/0.54/0.59
Volume fraction (%)	**Fuel**	34.26	29.30	22.08	17.44	16.0/20.6/26.1	20.4/26.2/33.3
	Bond	11.42	9.77	7.36	5.81	5.3/6.9/8.7	6.8/8.7/11.1
	Structure	25.73	25.68	26.41	29.15	28.5/30.4/31.4	31.4/31.4/32.4
	Coolant	28.59	35.25	44.15	47.60	50.1/42.1/33.9	41.4/33.6/23.2

[a] Inner core (IC)/middle core (MC)/outer core (OC).

[b] Primary/secondary control assemblies.

Table 3: Key core and fuel assembly design parameters of oxide fuel cores

Target TRU CR		**1.0**	**0.75**	**0.5**	**0.25**	**0.0**	
						MA/Pu ≅ 0.1	**MA/Pu ≅ 1.0**
Fuel assemblies		19/66/66[a]	72/36/36	72/36/36	72/36/36	78/24/42	78/24/42
Control assemblies		9/3[b]	16/3	16/3	22/3	22/3	22/3
Fuel pins per assembly		271	271	324	324	324/324/324	324/324/324
Fuel pin diameter (cm)		0.87	0.81	0.66	0.56	0.44/0.46/0.48	0.48/0.50/0.53
Volume fraction (%)	**Fuel**	49.29	41.65	30.22	19.73	10.4/11.9/13.5	13.2/15.1/17.2
	Bond	2.55	2.16	1.56	1.02	0.54/0.62/0.70	0.68/0.78/0.89
	Structure	28.58	27.71	29.22	26.22	22.8/23.4/24.0	23.9/24.7/25.4
	Coolant	19.58	28.48	39.00	53.02	66.4/64.1/61.8	62.2/59.4/56.5

[a] Inner core (IC)/middle core (MC)/outer core (OC).

[b] Primary/secondary control assemblies.

Fuel cycle performances

The equilibrium cycle core performance parameters obtained with the make-up TRU feeds of MA/Pu ≅ 0.1 and MA/Pu ≅ 1.0 are summarised in Tables 4 and 5. Because of the aforementioned design approach to control the burn-up reactivity swing with a moderate number of primary control assemblies, the cycle length decreases and the burn-up reactivity loss increases as the TRU conversion ratio decreases. Since it was maximised with the fast fluence limit of cladding, the fuel residence time shows relatively small variations with TRU conversion ratio. When the TRU conversion ratio is reduced from 1.0 to 0.0, the HM inventory is decreased by a factor of 4 to 5, and hence the discharge burn-up is increased by 2.5 to 4 times. On the other hand, the TRU inventory is increased by 10 to 90%, depending on the fuel type and make-up TRU feed stream. Compared to the metal cores, the oxide cores have larger HM and TRU inventories because of the higher active core height, which in turn allow longer cycle lengths. The fuel residence time also becomes longer because of the softer spectrum and reduced flux level (due to higher active core height).

Table 4: Equilibrium cycle performance parameters for make-up TRU feed of MA/Pu ≅ 0.1

Target TRU CR		1.0	0.75	0.5	0.25	0.0
Metal fuel core	Cycle length, days	370	232	221	158	132
	Average fuel residence time, days	1 351	1 448	1 380	1 293	1 284
	Average TRU enrichment, %	13.9	21.2	33.3	55.5	98.6
	TRU conversion ratio	1.001	0.749	0.502	0.245	0.004
	HM inventory at BOC, MT	16.75	13.44	9.45	5.86	3.62
	TRU inventory at BOC, MT	2.45	2.86	3.08	3.22	3.57
	Ave. discharge burn-up, MWd/kg	73	100	132	183	294
	Peak fast fluence, 10^{23} n/cm^2	4.00	3.86	4.00	4.07	4.00
	Burn-up reactivity loss, %Δk	-0.06	1.49	2.98	3.78	4.35
Oxide fuel core	Cycle length, days	607	353	326	165	124
	Average fuel residence time, days	2 216	2 204	2 039	1 851	1 582
	Average TRU enrichment, %	16.9	25.1	38.0	59.9	99.9
	TRU conversion ratio	1.001	0.753	0.499	0.250	0.001
	HM inventory at BOC, MT	19.28	15.25	10.91	6.86	3.82
	TRU inventory at BOC, MT	3.49	3.88	4.08	4.00	3.81
	Ave. discharge burn-up, MWd/kg	103	131	166	229	328
	Peak fast fluence, 10^{23} n/cm^2	4.00	4.00	3.91	4.00	4.10
	Burn-up reactivity loss, %Δk	0.20	1.80	3.57	3.02	4.06

Table 5: Equilibrium cycle performance parameters for make-up TRU feed of MA/Pu ≅ 1.0

Target TRU CR		1.0	0.75	0.5	0.25	0.0
Metal fuel core	Cycle length, days	370	232	221	158	132
	Average fuel residence time, days	1 351	1 448	1 380	1 293	1 284
	Average TRU enrichment, %	13.9	23.6	39.7	69.4	97.4
	TRU conversion ratio	1.015	0.722	0.442	0.163	0.008
	HM inventory at BOC, MT	16.75	13.44	9.46	5.88	4.82
	TRU inventory at BOC, MT	2.45	3.19	3.71	4.11	4.68
	Ave. discharge burn-up, MWd/kg	73	100	132	183	230
	Peak fast fluence, 10^{23} n/cm^2	4.00	4.05	4.26	4.28	4.02
	Burn-up reactivity loss, %Δk	-0.06	1.03	1.93	2.36	2.23
Oxide fuel core	Cycle length, days	607	353	326	165	124
	Average fuel residence time, days	2 216	2 204	2 039	1 851	1 582
	Average TRU enrichment, %	16.9	27.5	44.7	74.3	97.9
	TRU conversion ratio	1.022	0.727	0.435	0.156	0.007
	HM inventory at BOC, MT	19.28	15.26	10.93	6.88	5.12
	TRU inventory at BOC, MT	3.49	4.27	4.85	5.07	5.01
	Ave. discharge burn-up, MWd/kg	103	131	166	228	257
	Peak fast fluence, 10^{23} n/cm^2	4.00	4.32	4.29	4.37	4.17
	Burn-up reactivity loss, %Δk	0.20	1.30	2.30	1.86	2.00

For the make-up TRU feed of MA/Pu ≅ 0.1, the core-average TRU charge enrichments to achieve the target conversion ratios of 1.0, 0.75, 0.50, 0.25 and 0.0 in the metal fuel cores are about 14%, 21%, 33%, 56% and 99%, respectively. The average TRU enrichments to achieve the target conversion ratios are slightly higher for the oxide cores because of the softer spectrum and the less reactive equilibrium fuel compositions due to increased irradiation. Since the same assembly designs were used for both TRU feeds, except for the uranium-free fuel cores, the calculated conversion ratios for the make-up TRU feed of MA/Pu ≅ 1.0 are slightly different from the target values. With the make-up TRU feed of MA/Pu ≅ 1.0, the burn-up reactivity swing is reduced significantly; the degree of reduction increases with decreasing conversion ratio, up to 2 times for the uranium-free cores. On the other hand, because of reduced reactivity worth of the fuel material, the TRU inventory increases up to 30%.

Table 6 shows the minor actinide fraction of the equilibrium cycle charge TRU, and Table 7 presents the HM and TRU mass flows normalised to the same energy production, in the unit of effective full power year (EPFY). The minor actinide fraction in the equilibrium cycle charge TRU increases with decreasing TRU conversion ratio because of increased discharge burn-up and TRU enrichment. As the TRU conversion ratio decreases from 1.0 to 0.0, the heavy metal charge rate decreases by a factor of 2.5 to 4, depending on the fuel type and make-up TRU feed stream; due to the longer fuel residence time, the oxide cores need smaller HM charge rates than the metal cores. On the other hand, the TRU charge rate increases with decreasing TRU conversion ratio because of the increased TRU enrichment. The net TRU consumption rate increases from zero for the break-even cores to ~380 kg/EPFY for the zero conversion ratio cores.

Table 6: Minor actinide fraction of equilibrium cycle charge TRU

Target TRU CR		1.0	0.75	0.5	0.25	0.0
TRU feed of MA/Pu ≅ 0.1	Metal fuel	0.032	0.081	0.114	0.139	0.156
	Oxide fuel	0.044	0.089	0.119	0.140	0.156
TRU feed of MA/Pu ≅ 1.0	Metal fuel	0.032	0.222	0.332	0.404	0.434
	Oxide fuel	0.044	0.220	0.331	0.404	0.429

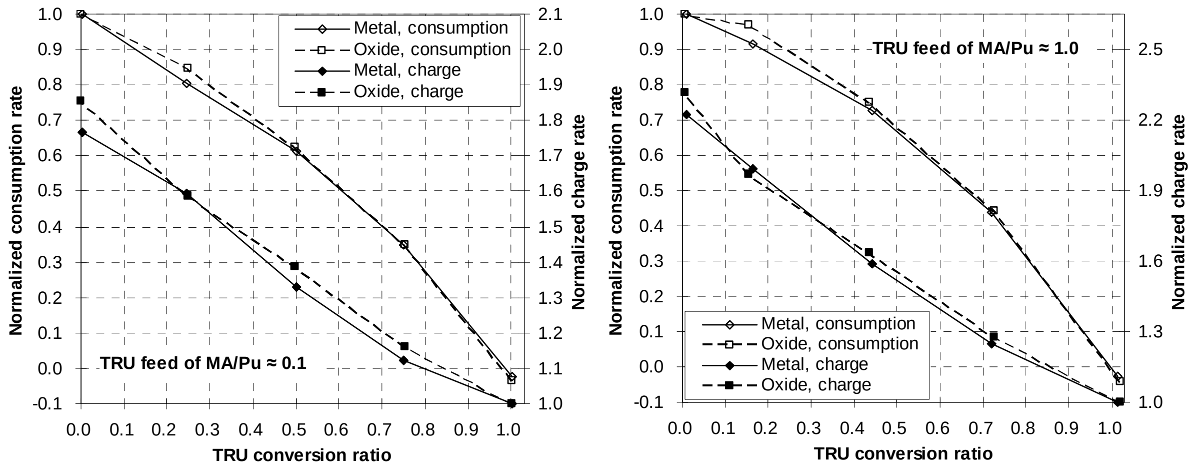
Table 7: Equilibrium cycle HM and TRU mass flows (kg/EFPY)

Target TRU CR				1.0	0.75	0.5	0.25	0.0
TRU feed of MA/Pu ≅ 0.1	Metal fuel	Charge	HM	4 847	3 555	2 683	1 931	1 204
			TRU	672	754	894	1 071	1 187
		Net TRU consumption		-5	83	174	273	376
	Oxide fuel	Charge	HM	3 440	2 700	2 129	1 542	1 080
			TRU	582	676	808	924	1 079
		Net TRU consumption		-7	83	178	274	379
TRU feed of MA/Pu ≅ 1.0	Metal fuel	Charge	HM	4 847	3 555	2 683	1 931	1 533
			TRU	672	840	1 066	1 340	1 493
		Net TRU consumption		-5	93	195	308	375
	Oxide fuel	Charge	HM	3 440	2 700	2 129	1 542	1 376
			TRU	582	743	951	1 146	1 347
		Net TRU consumption		-7	93	201	312	378

In order to reduce the legacy TRU inventory or to increase the support ratio to optimise the LWR to ABR ratio in the total nuclear fleet (under the assumption that ABR is less economical than LWR), it is desirable to reduce the TRU conversion ratio as low as practically possible within the safety and TRU fuel related constraints. With a low TRU conversion ratio, the amount of spent ABR fuel to be reprocessed is also reduced significantly due to the significantly smaller HM charge rate. On the other hand, the initial TRU inventory required to start the ABR and the TRU charge rate to produce the same amount of energy increase with decreasing TRU conversion ratio. If the fractional reprocessing loss is constant, the increase in the required amount of TRU would increase the reprocessing loss of TRU to the geological repository.

Figure 2 shows the TRU consumption rates relative to the maximum theoretical value of uranium-free fuel and the TRU charge rates relative to the break-even core as a function of TRU conversion ratio. The slope of TRU consumption rate with respect to the TRU conversion ratio decreases with decreasing TRU conversion ratio. The TRU consumption rate reaches ~80% of the maximum theoretical value when the TRU conversion ratio is in the range of 0.25-0.35. On the other hand, the slope of the TRU charge rate increases with decreasing TRU conversion ratio, more rapidly for the make-up TRU feed of MA/Pu ≅ 1.0. These results indicate that if two design objectives of maximising the TRU consumption rate and minimising the reprocessing loss of TRU to the geological repository are pursued, a compromise TRU conversion ratio would be in the range of 0.2 to 0.4.

Figure 2: Normalised TRU charge and consumption rates vs. TRU conversion ratio



Safety parameters

To investigate the feasibility of the critical cores corresponding to the different conversion ratios, the kinetics parameters and reactivity coefficients were calculated for the beginning of cycle (BOC) and end of cycle (EOC) configurations of the recycled equilibrium cores. Figure 3 presents the effective delayed neutron fraction, prompt neutron lifetime, Doppler constant, radial expansion coefficient, axial expansion coefficient, sodium density coefficient at EOC as a function of the TRU conversion ratio. The Doppler constant $A_D = T(\partial\rho/\partial T)$ is the proportional constant of the Doppler coefficient which is assumed inversely proportional to the absolute temperature. The radial expansion coefficient represents the reactivity effects of uniform, radial thermal expansion, and the axial expansion coefficient represents the reactivity effects of uniform, axial thermal expansion of fuel.

As shown in Tables 4 and 5, the TRU fraction in HM increases with decreasing TRU conversion ratio. Thus, the effective delayed neutron fraction decreases monotonically due to reduced ^{238}U fission. The reduced ^{238}U fraction also makes the Doppler constant less negative. It is noted that the Doppler constant of the uranium-free core (*i.e.* CR~1.0) fuelled with TRU feed of MA/Pu $\cong$ 1 is almost zero for the metal core and slightly positive for the oxide core. The HM loading decreases with decreasing TRU conversion ratio. The reduced absorption in HM increases the prompt neutron lifetime and makes both the radial and axial expansion coefficients more negative by increasing the leakage fraction. The sodium density coefficient generally becomes more positive with decreasing TRU conversion ratio since the positive spectral effect outweighs the negative leakage effect, but it does not show a monotonic behaviour because of the variations in the core configuration, material volume fractions and TRU inventory.

The integral reactivity parameters for the quasi-static reactivity balance analysis of the passive regulation of power [5,6] were estimated without considering the control rod driveline and vessel thermal expansion. The sufficient conditions for acceptable asymptotic core outlet temperatures for possible unprotected accident scenarios (loss of heat sink, transient overpower, loss of flow, chilled inlet and pump over speed) are:

$$A/B \le 1\,,\ 1 \le C\Delta T_c/B \le 2\,,\ \Delta\rho_{TOP}/|B| \le 1$$

The parameter *A* is the reactivity decrement due to the fuel temperature rise from the coolant average temperature to the fuel average temperature. The parameter *B* represents the reactivity decrement experienced when the coolant and fuel temperatures rise from the zero-power isothermal condition at coolant inlet temperature to the coolant average temperature. The parameter *C* is the reactivity decrement due to the coolant inlet temperature rise. The variable ΔT_c is the average coolant temperature rise across the core, and $\Delta\rho_{TOP}$ is the transient overpower (TOP) initiator. The ratios A/B and $C\Delta T_c/B$ at EOC are shown in Figure 4, and the maximum allowable reactivity fault of a single control assembly to satisfy the third condition is shown in Table 8.

Figure 3: Kinetics parameters and reactivity coefficients at EOC vs. TRU conversion ratio

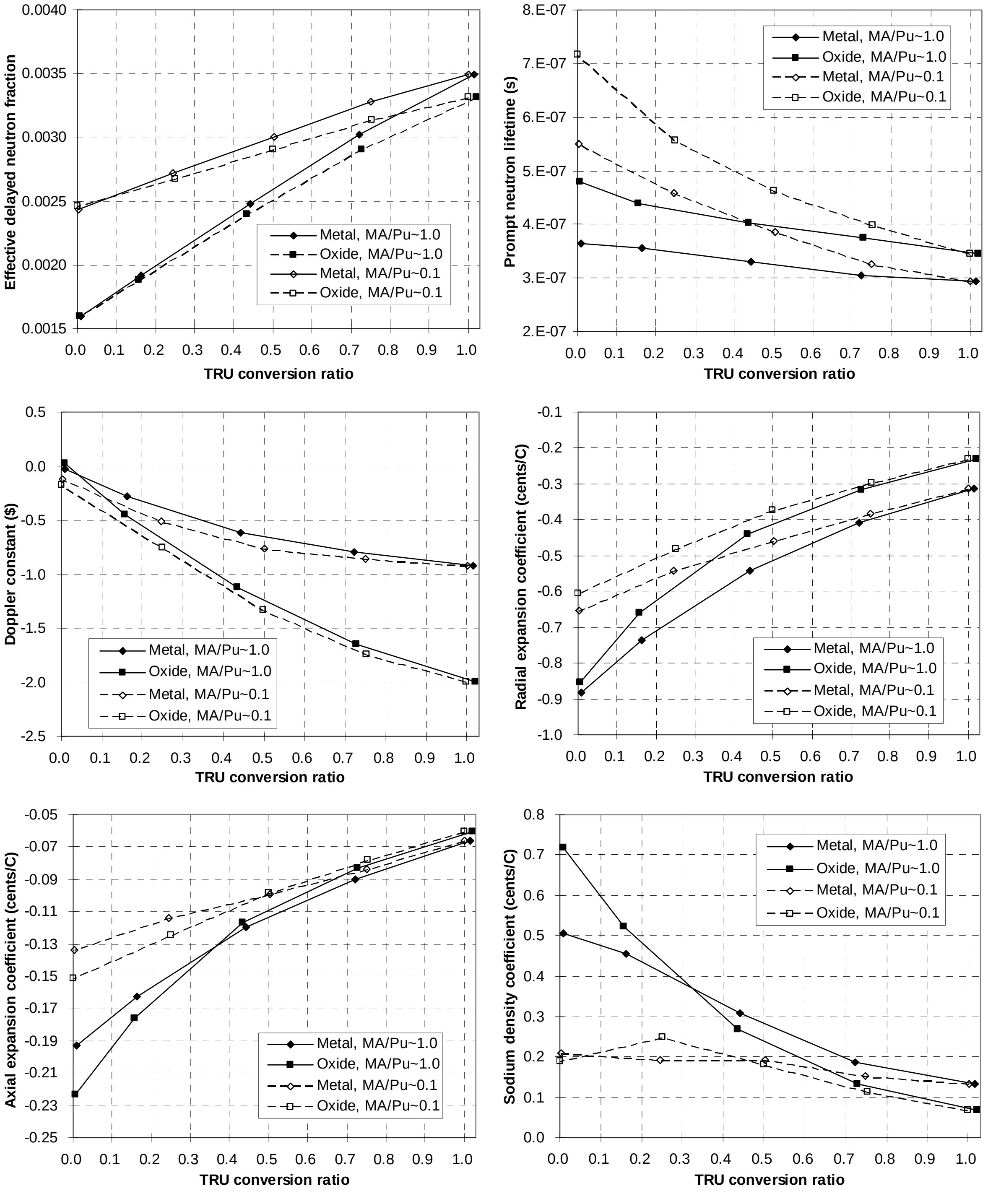


Figure 4: Integral reactivity parameter ratios at EOC vs. TRU conversion ratio

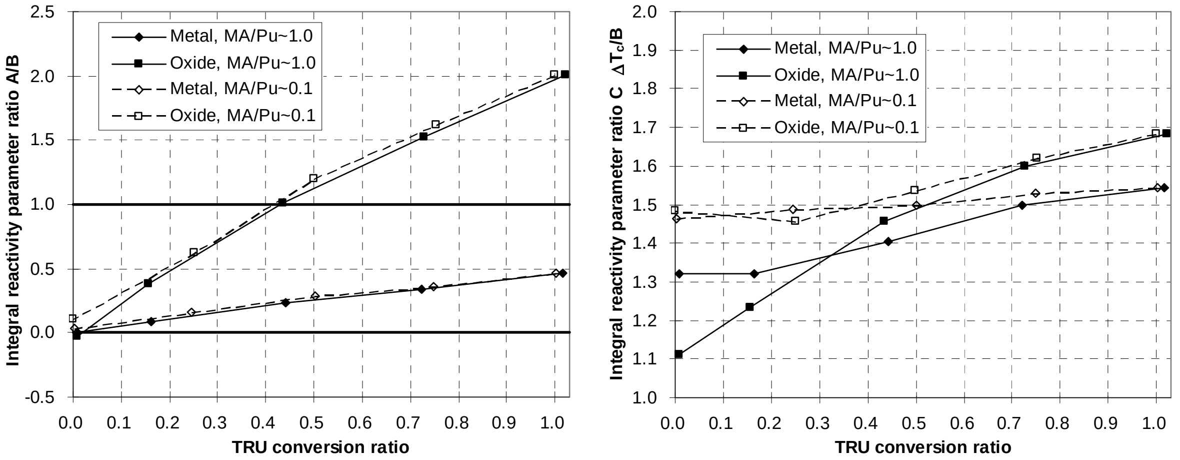


Table 8: Maximum reactivity fault (cents) of a single control assembly for passive shutdown

Target TRU CR		1.0	0.75	0.5	0.25	0.0
TRU feed of MA/Pu ≅ 0.1	Metal fuel	31	37	40	46	53
	Oxide fuel	33	35	35	39	52
TRU feed of MA/Pu ≅ 1.0	Metal fuel	31	37	40	47	57
	Oxide fuel	33	35	35	38	42

The calculated integral reactivity parameters showed the trend that as the TRU conversion ratio decreases, A becomes less negative and B becomes more negative. The parameter C becomes more negative with decreasing TRU conversion ratio, except for the oxide fuel cores with the TRU feed of MA/Pu ≅ 1.0, for which it showed an opposite trend. As a result, the ratios A/B and $C\Delta T_c/B$ decrease monotonically with decreasing TRU conversion ratio. As seen in Figure 4, the ratio $C\Delta T_c/B$ sufficiently satisfies the conditions for all the core designs of TRU conversion ratios from 1.0 to 0.0. For very small TRU conversion ratios, however, the ratio A/B becomes too small to satisfy the sufficient condition, mainly because of the diminished Doppler effects. In addition, the oxide cores do not satisfy this sufficient condition for conversion ratios higher than ~0.45, because of the large power coefficient A due to high fuel temperature and large Doppler coefficient. Additional design enhancements, such as a self-actuating shutdown device, gas expansion modules, extended pump coast-down, or low power density would be required for these oxide cores to increase safety margins so that no fuel damage or cladding failures would occur at the unprotected loss of flow event. As shown in Table 8, the maximum allowable reactivity fault of a single control assembly to satisfy the sufficient condition for the TOP passive shutdown increases with decreasing TRU conversion ratio. This trend mitigates the reactivity control burden for the increasing burn-up reactivity swing with decreasing TRU conversion ratio.

Conclusions

In order to generate representative data useful in selecting an optimum TRU conversion ratio for different technical options, a systematic study was performed on the trends in transmutation performance and safety parameters versus TRU conversion ratio for sodium-cooled recycle reactors. Using ternary metal and mixed-oxide fuels, ABR core concepts of 1 000 MWt power rating were developed for target TRU conversion ratios of 1.0, 0.75, 0.50, 0.25 and 0.0. Two make-up TRU feeds were considered with MA/Pu ratios of 0.1 and 1.0. Detailed fuel cycle analysis was performed and TRU consumption rate and isotopic mass flow rates were evaluated. Kinetics parameters and reactivity feedback coefficients were also calculated, and the integral reactivity parameters for the quasi-static reactivity balance analysis of the passive regulation of power were estimated.

As the TRU conversion ratio decreases, the initial heavy metal inventory and charge rate decrease by a factor of 3 to 5, depending on the fuel type and make-up TRU feed stream. On the other hand, the TRU inventory and charge rate increases, but to a lesser extent. With decreasing TRU conversion ratio, the slope of TRU consumption rate with respect to the TRU conversion ratio decreases but the slope of the TRU charge rate increases. The TRU consumption rate reaches ~80% of the maximum theoretical value when the TRU conversion ratio is in the range of 0.25-0.35. These results indicate that if two design objectives of maximising the TRU consumption rate and minimising the reprocessing loss of TRU to the geological repository are pursued, a compromise TRU conversion ratio would be in the range of 0.2 to 0.4. The estimated safety parameters indicate that the metal and oxide cores of TRU conversion ratio in the range of 0.25-0.40 are feasible with favourable passive safety features. The safety parameters of these cores are comparable to those of conventional fast reactor cores that have already been proven to be feasible, and the sufficient conditions for acceptable asymptotic core outlet temperatures for possible unprotected accident scenarios are satisfied.

References

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The effectiveness of the ELSY concept with respect to minor actinide transmutation capability

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Abstract

The task of partitioning and transmutation (PT) aims at the sustainability of new global nuclear scenarios for energy production, required by a continuously growing demand.

The nuclear renaissance boosted by the breaking need of a reduction in CO_2 *emissions, together with increasing safety and security requirements, is creating a clear interest in the Generation-IV philosophy. In particular, an effective management of minor actinides (MA) and their multi-recycling in innovative fast spectrum systems can lead to a minimisation of high-level wastes (HLW) to be disposed of in geological repositories.*

This study presents a PT application based on the European Lead-cooled System (ELSY), the 600 MWe Gen-IV lead-cooled fast reactor (LFR) under investigation in Europe within the 6th EURATOM Framework Programme. An "adiabatic" core configuration is investigated here, for a system which can maintain a constant amount of both MA and plutonium during the whole fuel cycle, even without either axial or radial blankets.

It is shown that an equilibrium concentration of MA exists, for which its production rate is exactly compensated by its transmutation rate. Any other concentration may enhance either their production or removal in such a way as to allow the system to evolve almost exponentially towards the equilibrium state.

The practical feasibility of such an equilibrium core is then analysed: acceptable operative conditions might lead to a "sustainable" nuclear system, the overall net outcome of which is the production of energy by burning a feed stream of uranium from one side, and the discharge of fission products (FP) only from the other side.

Introduction

The management of the spent fuel discharged from nuclear reactors is one of the most demanding issues for the overall sustainability of the whole nuclear fuel cycle. Actually, the presence of long-lived radioisotopes (LLR) in the discharged fuel, and in particular of minor actinides (MA), requires the adoption of partitioning and transmutation (PT) strategies intended to minimise the amount of MA in the final wastes.

At first MA have to be separated from the spent fuel matrix in order to be further processed or separately stored: the obtained MA-free matrix, containing almost only fission products (FP), even if highly radioactive can therefore be stored in geological repositories for human affordable time lapses (some hundred years). The pursued sustainability of the nuclear fuel cycle can then be finally achieved by multi-recycling the MA in innovative fast spectrum systems, using U-Pu matrix as nuclear fuel.

In particular, the MA management proposed in this paper consists in recycling the MA in the European Lead-cooled System (ELSY) [1,2], in such a way as to maintain a constant amount of both MA and Pu by equilibrating their disappearance and their production along the cycle. The equilibrium concentration of transuranics (TRU) in the MOX matrix characterises the so-called "adiabatic" core concept: a nuclear reactor the net outcome of which would only be the transmutation of a feed input stream of depleted uranium (DU) into FP.

The "adiabatic" core concept

The MA burning in a fast reactor is the result of the fissioning of the target nuclides by the neutron flux corresponding to a given power level. On the other hand, the Pu sustaining the chain reaction and the fertile U are at the same time transmuted into MA and (mainly) Pu respectively.

The build-up of each isotope, as well as its depletion, depends on the neutron spectrum of the reactor which determines the effective absorption and fission cross-sections. In a fast reactor fuelled with fresh pure MOX, the build-up of the MA (initially absent) follows a non-linear behaviour. In fact, as far as MA are produced in the core, they are subject to the same neutron flux, causing their fission and a partial balancing of their build-up. On the other hand, it has been proven [3] that the burning of a MOX fuel highly enriched in MA results in a net depletion of such isotopes, that is, the burn-up (BU) of these isotopes exceeds their build-up.

The MA time evolution depends on their concentration in the fuel: the richer the fuel in MA, the more actinides are burned rather than produced, and *vice versa*. The concentration of each isotope will then follow an exponential evolution (depletion or build-up, depending on the initial concentration of the isotope itself and of its parent, respectively) towards an equilibrium concentration, which is univocally fixed by the cross-sections and the neutron spectrum. The time constant of these concurrent mechanisms is proportional to the neutron flux.

The adiabatic core concept is based on this mechanism: if the fuel of a fast reactor contains exactly the equilibrium concentration (which is a characteristic of each core via spectrum) of MA and Pu, then the system will maintain unaltered that amount during the whole fuel cycle, by exactly compensating transmutation and fission reaction rates. Such a system will therefore burn its own MA, resulting in a sort of black-box converting DU into FP only.

The ELSY reactor

The reactor which has been considered in the present work to demonstrate the viability of the adiabatic core concept is ELSY, whose neutronics is under investigation at the Bologna ENEA research centre. ELSY is a pool-type lead-cooled 600 MWe fast reactor, under development within the 6th EURATOM Framework Programme. In particular, ELSY aims at fulfilling the requirements of sustainability, proliferation resistance, physical protection, safety and economics, and represents a European solution for the lead-cooled fast reactor (LFR), one of the six most promising nuclear systems proposed by the Generation-IV International Forum [4].

The neutronic characteristics (hard spectrum, criticality swing in the cycle and optimal breeding ratio) obtained by the calculations performed so far on a preliminary MA-free ELSY core [5], mesh well with the aimed task.

Core configuration

The core configuration considered in the present work is the so-called "open square", characterised by traditional PWR-like fuel assemblies (FA) made up of a wrapper-less square 17 × 17 pin lattice, as presented in [5]. The alternative solution is a classical fast reactor core composed of hexagonal FA with wrapper [6].

The FA developed at ENEA is made of 284 fuel rods and 5 structural pins (located at the 4 corners and in the central position), as shown in Figure 1.

The criticality of the core is reached by arranging 272 FA in a pseudo-cylindrical scheme of about 5 m diameter (Figure 2).

Figure 1: Horizontal cross-section of the ELSY open-square FA

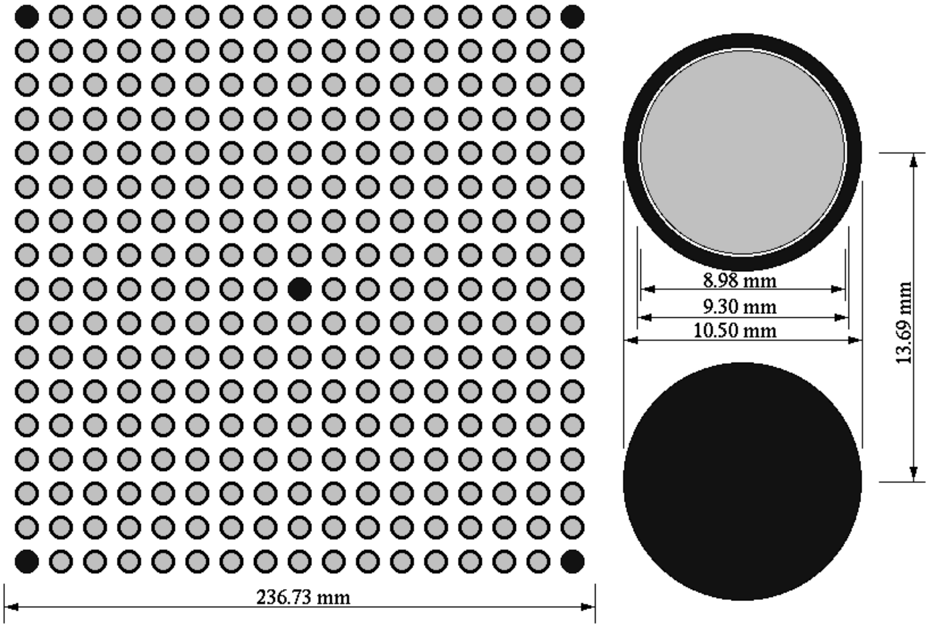
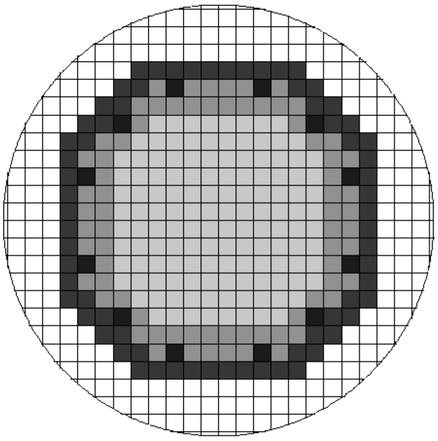


Figure 2: Horizontal cross-section of the open square ELSY core



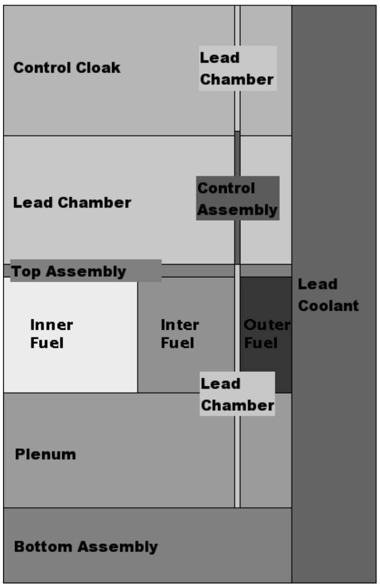
Since the FA are open, there is no possibility of gagging the FA in order to tune the Pb flow rate in each element so as to achieve the same output temperature, for a better efficiency of the system. The only possible alternative is to maintain the FA power distribution as flat as possible by segmenting the core into three concentric regions with different Pu contents (different grey tones in Figure 2), namely 13.4, 15.0 and 18.5$^{v}/_{0}$ (volumetric per cent) of $PuO_{1.95}$, obtaining an overall maximum-to-average assembly power distribution factor of 1.15.

The 12 assembly positions marked in black in Figure 2 represent the control rods (CR) [5] as homogeneous media made up for a 70$^{v}/_{0}$ of 90$^{a}/_{0}$ (atomic per cent) B^{10} enriched B_4C.

The 90 cm high active region and the adjacent structural elements have been simulated with a simplified zoned model of the ELSY core (see Figure 3), according to the capabilities of the ERANOS [7] deterministic code used for the simulations.

A more detailed description (in terms of both geometry and compositions) of the adopted core model is presented in Ref. [5].

Figure 3: Axial cross-section of the ELSY core model considered in the present work



Preliminary burn-up evaluation

A preliminary BU analysis performed on the ELSY cycle [5] has provided the basic data needed to investigate the MA concentration evolution, starting from beginning of life (BoL) fresh fuel. The results, plotted in Figure 4, show the typical exponential behavior.

Using simple calculations based on the numerical values of Figure 4, it has been possible to retrieve the parameters characterising, for each element, the build-up law, assumed to be of the form:

$$C(t) = C_0\left(1 - e^{-\tau \cdot t}\right)$$

where the concentrations [both the equilibrium one, C_0, and the time-dependent one, $C(t)$] are expressed as mass fractions of the total mass of heavy metals (HM) present in the fuel, while the time constant τ is expressed in y^{-1}.

The first trial C_0 and τ parameters for the MA elements and for each fuel zone are summarised in Table 1. It is worth noting that only the estimates for Am and Np can be considered reliable, since their evolution mainly depends directly on pre-existing isotopes; on the other hand, the evolution of Cm is strictly dependent on the build-up of Am, and therefore cannot be considered for equilibrium calculations. This concept is similar to the secular equilibrium of radioactive isotopes in the natural decay chain, where the activity of an isotope depends on the activity of its parent.

Figure 4: The time behavior of the MA element concentrations

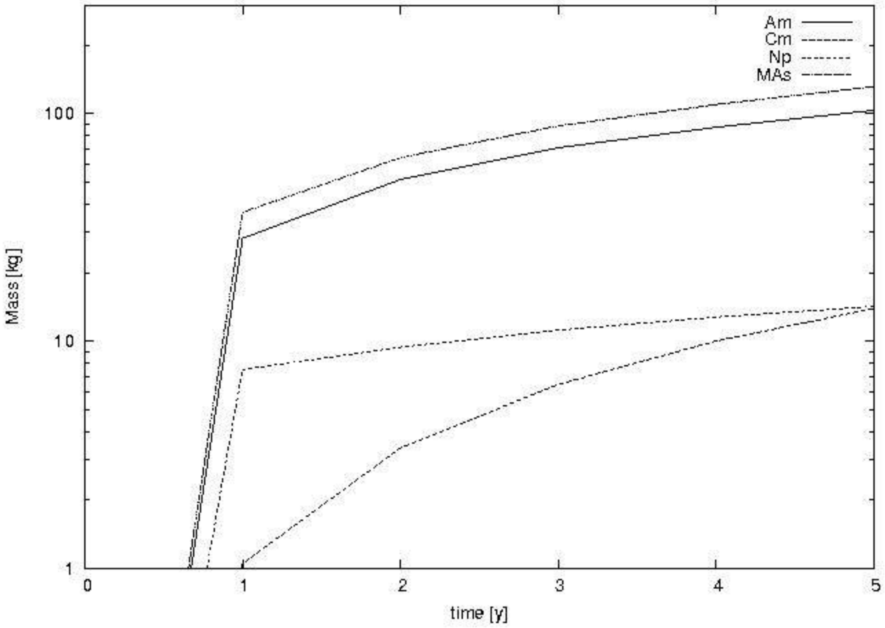


Table 1: Equilibrium concentrations and time constants for the MA in the different fuel zones of the ELSY core

	Inner fuel zone		Intermediate fuel zone		Outer fuel zone	
	C_0	τ	C_0	τ	C_0	τ
Am	5.09E-03	0.145	5.71E-03	0.127	1.07E-02	0.065
Cm	8.77E-03	0.012	3.78E-03	0.016	1.30E-03	0.029
Np	5.24E-04	0.435	3.42E-04	0.550	2.22E-04	0.574

The parameters of the previous table, together with the mass equivalence coefficients, have been used to calculate the masses of Pu and MA to be loaded at BOL in ELSY, for the first simulation of the adiabatic fuel cycle. The only constraints of this work scheme are the vectors of input actinides to be used (presented in Table 2), which fix the isotopic composition of each element.

Table 2: Isotopic compositions of reactor-grade Pu, depleted U, Am and Cm vectors

Plutonium	
Isotope	[$^w/_0$]
^{238}Pu	2.333
^{239}Pu	56.873
^{240}Pu	26.997
^{241}Pu	6.104
^{242}Pu	7.693

Uranium	
Isotope	[$^w/_0$]
^{234}U	0.003
^{235}U	0.404
^{236}U	0.01
^{238}U	99.583

Americium	
Isotope	[$^w/_0$]
^{241}Am	82.118
^{242f}Am	0
^{242m}Am	0.277
^{243}Am	17.605

Curium	
Isotope	[$^w/_0$]
^{243}Cm	1.533
^{244}Cm	69.763
^{245}Cm	26.588
^{246}Cm	2.076
^{247}Cm	0.039

The equilibrium fuel cycle

A complete model of the ELSY reference core has been implemented in the ERANOS [7] deterministic code. MOX fuel with the theoretical concentrations of MA calculated as above has been considered for a first trial simulation of the fuel cycle. An iterative refinement of the equilibrium concentrations evaluation has to be performed, since the amount of MA in the core alters both the spectrum and the breeding of the system, and thus the MA concentration evolution itself.

The reliable ERALIB1 cross-section library has been chosen for both the cell calculations and the BU simulation. This data library has been derived from the JEF2.2 [8] point-wise one, updated, for the most important nuclides, by means of integral data obtained at the MASURCA facility [9].

Transport calculations (in P1 model) have been performed for the cells describing the reactor, by means of the European Cell Code (ECCO) [10], in order to retrieve the effective cross-section data sets at 33 energy groups needed for the core simulation. The latter has been performed by means of the ERANOS 2-D diffusion module BISTRO [11].

The equilibrium concentration

The initial compositions for the three fuel regions have been recalculated after each simulation by considering the ^{239}Pu equivalent masses of the MA isotopes, so as to maintain the criticality of the system and the FA power distribution factor.

The results presented in Table 3 indicate that, despite the fixed MA vectors, the equilibrium concentrations can be achieved in the ELSY core by means of acceptable amounts of MA in the fresh fuel, without affecting either the breeding or the kinetic parameters of the reactor [12]. As expected, the isotopic composition of the elements vectors exhibit a rearrangement coherent with the different isotopes' cross-sections.

Table 3: MA equilibrium concentration in the three ELSY fuel regions

	INN	INT	OUT
Am	5.18E-03	6.95E-03	1.29E-02
Cm	3.05E-03	2.23E-03	9.98E-04
Np	9.03E-04	1.07E-03	7.80E-04

The actual ELSY fuel cycle

The mean residence time of each FA in the ELSY core is fixed to four years, with a refuelling strategy of one quarter of every core region each year: every FA is then loaded at time 0, and is removed at time 4. The BU of the fuel in the core is therefore rather inhomogeneous, as well as its isotopic composition. Moreover, the partially burned FA are rearranged within the different regions in order to achieve a BU as flat as possible and to mitigate the consequences of the radial neutron flux gradient on the system performances.

On the other hand, the ERANOS simulations of the ELSY fuel BU are performed on an initially fresh fuel, uniformly loaded in the core, and at fixed time intervals.

The classical solution of this problem is to consider the composition of the whole core in first approximation as a homogeneous mixing of 0-, 1-, 2- and 3-year burned fuel at beginning of cycle (BOC), and of 1-, 2-, 3- and 4-year burned fuel at the end of cycle (EOC). Therefore, the mean ageing of the fuel is 2.5 y and 1.5 y just before and immediately after the refuelling, respectively.

Under this hypothesis, the satisfactory results obtained for a uniformly burned fuel and presented in the previous subsection are confirmed: as shown in Table 4, MA content in the core is almost constant along the fuel cycle. This demonstrates the viability of the "adiabatic" core concept.

Table 4: MA evolution along the ELSY fuel cycle

	Mass at BOC [kg]	Mass at EOC [kg]	Relative variation [%]
Am	2.91E+02	2.89E+02	-0.79
Cm	1.13E+02	1.12E+02	-0.96
Np	3.88E+01	3.87E+01	-0.14

The criticality of the system and the reactivity swing during the fuel cycle also confirm the viability of this solution; actually, the BOC k_{eff} is 1.00415, and the reactivity swing for the whole cycle is 358 pcm.

Concluding remarks

A particular application of a partitioning and transmutation strategy which foresees MA recycling in the European LFR ELSY has been presented.

In particular, it the viability of implementing in ELSY the so-called "adiabatic" core concept has been proven, by identifying the equilibrium concentrations for each MA, and then simulating a realistic operating four-year long fuel cycle, with the substitution of one-quarter of the whole core every yearly sub-cycle.

Due to the constraint of fixed MA isotopic composition, the physical equilibrium of each isotope has not been reached. On the other hand, it has been possible to define the initial concentrations of MA which allow maintaining their total mass in the core constant along the fuel cycle, *i.e.* with just an isotopic rearrangement in the vectors, with no net production of MA during the whole fuel cycle.

Acknowledgements

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Design, safety and fuel developments for the EFIT accelerator-driven system with CERCER and CERMET cores

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Abstract

European R&D for ADS design, fuel and general technology development is driven in the 6th FP of the EU by the EUROTRANS programme. In EUROTRANS, two ADS design routes are followed, the XT-ADS and the EFIT. The XT-ADS is designed to provide the experimental demonstration of transmutation in an accelerator-driven Ssstem. The longer-term EFIT development, the European Facility for Industrial Transmutation, with which this paper is deals, aims at a generic conceptual design of a full transmuter. The EUROTRANS Domain DM1 (DESIGN) developed the conceptual reference design of the EFIT, a 400 MW$_{th}$ *ADT, loaded with a CERCER, U-free fuel based on an MgO matrix. The Domain DM3 (AFTRA), responsible for fuel development within EUROTRANS, in parallel developed a core loaded with a Mo-92 CERMET matrix-based fuel. In both cores for the cladding, the 9Cr1MoVNb T91 steel has been chosen. The core coolant is pure lead with inlet and outlet temperatures of 673 K and 753 K. The windowless target for the 800 MeV proton beam also contains pure lead. The EFIT concept was intended to be optimised towards: a good transmutation efficiency, high burn-up, low reactivity swing, low power peaking, adequate subcriticality, reasonable beam requirements and a high level of safety. In the current paper the two designs are reported and discussed. For the project, detailed design calculations have been performed both with deterministic and Monte Carlo methods. An extensive safety study is currently under way for the CERCER reference core. For the CERMET core the most important safety analyses have already been performed, sufficient for a preliminary safety assessment. The status of the design work and fuel development for the CERCER and CERMET cores is presented. In addition the results of the CERMET safety analyses are given.*

1 Introduction

The 6th EU FP EUROTRANS programme [1], which drives the European R&D for ADS design, fuel and technology development develops two ADS designs, the XT-ADS and the EFIT. The XT-ADS is designed to provide the experimental demonstration of transmutation in an accelerator-driven system. The longer-term EFIT development, the European Facility for Industrial Transmutation, worked out to a high level of detail [2], aims at a generic conceptual design of a full transmuter. In the current paper only the status of the EFIT development is described and discussed. The EUROTRANS Domain DM1 (DESIGN) is responsible for the development of the conceptual reference design of the EFIT, a 400 MW_{th} ADT. This design is loaded with MA and Pu in a CERCER U-free fuel with a MgO matrix. The core coolant, allowing a fast spectrum, is pure lead, as well as the windowless target for the 800 MeV proton beam. The reference subcritical level has been postulated to be $k_{eff} = 0.97$, a figure that has to be confirmed by the full safety analysis.

The EUROTRANS Domain DM3 (AFTRA) has been responsible for the fuel development within EUROTRANS. To assess the different fuel forms and matrices also in AFTRA various cores have been developed both with composite fuels as CERCER and CERMET and also cores with solid solution fuels. Because of the vast experience existing on oxide fuels in Europe, the main emphasis of the ADT fuel development concentrated on the oxide route. The final recommendation of the AFTRA Domain on fuels gave a ranking of these fuels based on a number of criteria, ranging from fabrication, reprocessing via economics to safety. The composite CERMET fuel $(Pu_{0.5},Am_{0.5})O_{2-x}$ – ^{92}Mo (93% enriched) has been recommended by AFTRA as the primary candidate for the EFIT [3]. This CERMET fuel fulfils adopted criteria for fabrication and reprocessing, and provides excellent safety margins. Disadvantages include the cost for enrichment of ^{92}Mo and a lower specific transmutation rate of minor actinides. The composite CERCER fuel $(Pu_{0.4},Am_{0.6})O_{2-x}$ – MgO has been recommended as a back-up solution as it might offer a higher consumption rate of minor actinides, and can be manufactured for a lower unit cost. Because of the supposed superior transmutation performance of the CERCER, the DM1 design work concentrated on the CERCER core. The detailed neutronic design calculations have been performed both with the deterministic code ERANOS [4] and Monte Carlo methods. The Monte Carlo code MCNPX [5] has been used to calculate, starting from the 800 MeV proton, the neutron source for ERANOS. For the CERMET core ERANOS [4], DANTSYS [6], SIMMER-III [7-9] and C4P-TRAIN [10] have been used for the core design, the transmutation optimisation and the burn-up behaviour optimisation. Thermo-hydraulic analyses of the CERCER core were mostly done by means of the SIM-ADS code [11]. The thermo-hydraulics for the CERMET core has been performed with the SDP-C3 [12] and SIMMER-III codes [7,8].

Current analyses show that the CERMET variant can achieve a similar transmutation performance, mainly because of the larger flexibility in pin design [13]. An important advantage of the CERMET fuel lies in its high thermal conductivity. Thus one might *e.g.* increase the pin diameter or might upgrade the core to higher power densities and higher ratings. Scenario analyses on integrating the EFIT into efficient transmutation strategies showed that this could be a point of interest [14].

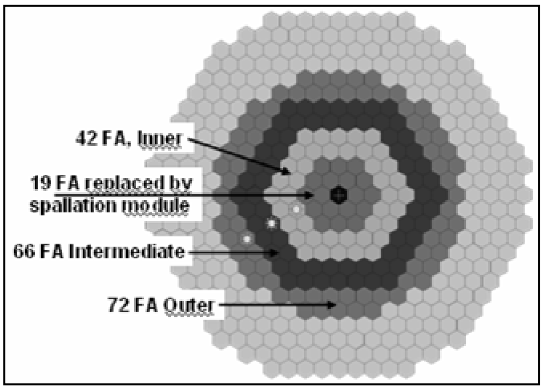
The EFIT designed by DM1 [2] represents an efficient "MA burner" where new MA are used for refuelling and only fission products are unloaded. Preliminary analyses show that this is possible with the EFIT with an equilibrium composition in which the equilibrium vector of the plutonium is however quite different from the initial one. The CERCER EFIT start-up core has been designed to fit the so-called "42:0" approach [2,15]. The fission rate is ~42 kg/TWh_{th}, representing the 200 MeV/fission in changed units of measure, in any nuclear system. The 42 kg/TWh_{th} is not a result of a design, but a physical constant. The "42" fissioned can however be differently split between MA and other heavy nuclides as Pu. Along the fissions (the universal 42), events on MA other than fission can occur. Thus, the MA "disappearing" can be actually higher than 42, that in turn would simply mean the exceeding part has been transmuted into other heavy nuclides (*i.e.* Pu). One can condensate all that in a pair of numbers: the first one indicates the overall MA disappearance (either fissioned or transmuted), the second one is an indicator of new Pu production, with their difference being in any case 42. The EFIT fundamental choice of the inert matrix implies that new Pu production has to be avoided. On the other hand the Pu fission, leading to its net decrease, would be significantly more expensive in an ADS than in a critical reactor. Therefore the Pu balance should be 0, that leads to the "42:0" pair. Of course it does not mean that every MA atom belonging to the "disappeared 42" is directly fissioned: a good part is transmuted into Pu and in the meantime an equal amount of Pu is fissioned. This has been accomplished with a fuel enrichment of Pu/(Pu + MA) = 45.7%. Should a different Pu policy be chosen, either Pu burning (< 42 for MA) or Pu producing (> 42 for MA), it would be possible to fit the design for this policy, however, the possible gains (in breeding/burning Pu) are limited by constraints related to core criticality and reactivity. It is important to note that, with the Pu content rather constant over the cycle, the reactivity swing will not be large. A low reactivity swing is of importance as the EFIT will be designed with a non-variable beam power.

The safety objectives common to all approaches for nuclear plants, including ADT, are that all reasonably practicable measures are taken to prevent accidents in nuclear installations, and to mitigate their consequences. This is achieved through the application of the defence-in-depth concept. The demonstration of the adequacy of design with the safety objectives is structured along three kinds of basic conditions: design basis conditions (DBC, structured in four categories), design extension conditions (DEC – limiting events, complex sequences and severe accidents) and residual risk situations [16]. For innovative reactors such as the ADT cliff-edge effects should be identified and excluded [17]. The fuel limits under the various accident categories have been specified by AFTRA. The in-pile experiments planned within AFTRA, FUTURIX [18] will provide information on the irradiation behaviour of CERCER and CERMET fuels. Information on He release will be provided by the HELIOS [19] and BODEX [20] experiments. As this information will become available only towards the end of the AFTRA project, the temperature limits are conservative and may be updated. Besides the fuel limits, clad limits have to be defined for the safety analyses. For the CERMET core the SIMMER-III code has been used up to now [21] as the main tool for the safety analyses and some new results will be reported here. Representative transients have been analysed as the unprotected loss of flow (ULOF), the unprotected over power (UTOP) and the unprotected blockage accident (UBA) and others. The transients give a good indication of critical issues of the ADS safety. A major point was the understanding of the accident scenario under a UBA accident as the total UBA leads to pin failures and some damage propagation potential. An important safety parameter in this respect is the significant positive void worth of the CERCER and CERMET cores and the strong production of He via the transmutation process. While coolant boiling can be excluded because of the high boiling point of lead coolant, pin failures could lead to a gas blow-down from the plena, to local voiding and reactivity addition. Another route to voiding may be the ingress of steam via a steam generator tube rupture accident (SGTR). Currently analyses for the latter scenario with the SIMMER code [22] do not indicate a rapid and large-scale voiding potential. In the section on safety below, results are shown for the pin failure scenario and a safety assessment is given. Safety is a major issue for cores having a high content of Am in their fuels.

2 The CERCER reference core design

In a contribution for the PHYSOR 2008 conference [2] a detailed description of the EFIT CERCER three-zone core with MgO as inert matrix is given. So only some key issues will be repeated here. The important decision in designing the core has been the definition of a unique enrichment that fits the "42:0" approach. The Pu content is rather constant in the cycle, resulting in a low reactivity swing which allows maintaining a rather constant proton current. A BU swing of only 200 pcm/year has been achieved. Correspondingly a rather constant current, of only about 15 mA, is required. The isotopic compositions of the used Pu and MA have been obtained as a result of a mixing of MA coming from the spent UO_2 fuel (90%) and the spent MOX (10%) of a typical PWR unloaded at a burn-up of 45 MWd/kgHM, then cooled down for a period of 30 years. Plutonium is extracted from the same spent UO_2 but with a storage period of 15 years. With these vectors the enrichment fitting the pair goal "42:0" has been evaluated and found to be 45.7%. The maximum linear power turns out to be 203 W/cm in the inner zone (close to the spallation source) and the average homogeneous power density is 70.7 MW/m^3.The power deposition in the target will not exceed 9 MW. The spread of the outlet temperatures of the subassemblies, belonging to the same zone of flow rate, must have a low peak factor (lower than 1.2 as a first approximation). In order to flatten the radial flux profile, the active fuel volume fraction is increased along the radius. As the "42:0" approach defines the enrichment, to flatten the radial flux profile the active fuel volume fraction has been increased along the radius. That has been achieved by either increasing the fuel/matrix ratio keeping the same pin diameter (from inner to intermediate zones), or by increasing the pin diameter keeping the same fuel/matrix ratio (from intermediate to outer zones). In the operating conditions, the mean outlet temperature of the coolant (pure lead) of 753 K is rather close to the maximum allowed temperature of the cladding of 823 K [23] and the maximal fuel temperatures T_{fuel} ~ 1 650 K. The chosen structural material is ferritic-martensitic steel T91, for which a maximum design temperature is allowed for the clad, taken into account a suitable treatment, is 823 K. For the EFIT clad and structural materials an aluminisation treatment with a GESA type technique is foreseen [24], so no thermal conductivity reducing oxidation layers have to be taken into account on the cladding surface. The impact of in-clad corrosion under burn-up conditions has been noted [21], but up to now not investigated in detail. At present a residence time of three years is foreseen for the fuel with a peak burn-up of about 8%, well below the dpa limit. The core has a HM mass of 5 350 kg. As a consequence of the selected enrichment, 45.7% ("42:0" approach), the mass of the Pu (some 2 450 kg) remains rather constant and supplies reactivity, while only the MA are fissioned. Over a three-year period in total 470 kg of MA are fissioned. The neutron source efficiency is rather low $\varphi^* = 0.52$, while in the PDS-XADS design [24] was $\varphi^* = 0.99$ (k_S and k_{eff} very similar). This effect is mainly due to the different fuel composition and to the larger radius of the target. The pin and pellet dimensions in the first two core zones are 8.62 versus 7.1 mm with a matrix fraction of 57% and 50%. In the outer zone the pin diameter is 9.52 and pellet is 8.0 mm again with a matrix fraction of 50%. The p/d ratio for the first two zones is thus 1.58 and 1.42 for the outer. Assembly outer and inner widths are 191 mm versus 178 mm with 168 fuel and 1 steel pin. While the pin diameter and the pitch derive from the thermal balance, the fuel assembly dimensions are driven by the size of the spallation module, which has to be inserted replacing the 19 central assemblies. The core is shown in Figure 2.1.

Figure 2.1: The three-zone EFIT core (180 fuel assemblies)



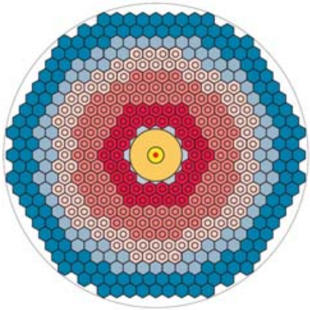
As described earlier, the core has been designed mainly by means of the deterministic code ERANOS [4] and the Monte Carlo code MCNPX [5]. The neutron libraries used for the codes are: ERALIB1 (JEF-2.2) for ERANOS; a combination of JEFF-3.1, ENDF/B-VI, LA150 [26] for MCNPX. For high energy interactions, the CEM03 physics model [5] has been used.

A key issue for the safety assessment is the calculated safety coefficients. The β_{eff} has been evaluated with 148 pcm for the BOC core. The core void effect, when the whole active core is voided is in the range of 6 684 pcm for the BOL and 6 666 pcm for the BOC core. Adding the plenum in the void consideration reduces the void worth by approximately 1 000 pcm. The void worths are therefore about twice the subcriticality value. Large-scale voiding by boiling can however be excluded for the HLM conditions.

3 The AFTRA CERMET core design

In the framework of EUROTRANS the AFTRA Domain developed various proposals for EFIT single-zone, two-zone and three-zone CERCER and CERMET cores [27]. The design studies for AFTRA were mainly performed with the ERANOS code system [4]. The EFIT cores were designed with the purpose to test and categorise the fuels which have been proposed within the AFTRA Domain. Therefore the cores were not optimised *e.g.* for achieving a maximum in transmutation efficiency. Issues like reducing the maximum void worth or going to higher power densities played a major role. EFIT cores with high-power densities were especially analysed to obtain phenomenological information in the high-temperature range and identify boundaries and margins. The results have been published [3,26,28,29] in various papers. Based on this premise it was shown that the MgO CERCER might be advantageous for achieving a higher transmutation rate for MA in case of the same pin/SA geometry, same Pu/MA ratio and criticality level. However, much better thermal physical properties of CERMET make a design with thicker pins (corresponding to a higher fuel volume fraction) possible to improve the transmutation efficiency without loss of any safety advantages. Therefore recently AFTRA has concentrated on developing a CERMET core along these lines. A three-zone core design was carried out, which satisfies the EFIT general and specific requirements and is adapted to the EFIT overall plant design [30,2]. To flatten the radial power profiles the "variable matrix technique" is applied in the first step with an upper limit of the fuel with 50 vol.%. The "42:0" condition has been relaxed for obtaining a good MA transmutation rate and achieving a low reactivity swing. The number of subassemblies and the associated fuel/matrix ratios in three zones are determined for a targeted subcritical level at k_{eff} about 0.97 and a required total form factor around 1.44. The core is displayed in Figure 3.1.

Figure 3.1: Cross plane of current AFTRA three-zone core with CERMET fuel



As already discussed, the Pu/MA ratio can be varied under the constraint of the "42" fission rate, also reducing a possible reactivity swing. Reducing the Pu/MA ratio causes a decrease of subcriticality. To maintain the same subcriticality, a higher fuel volume fraction is needed, provided that the reactor dimensions have to be kept constant. Therefore the pin diameter has to be increased. A thicker pin leads to a harder neutron spectrum, and thus to a better transmutation performance. Simply increasing the pin diameter will weaken the thermal performance of the core, *i.e.* causing the coolant volume fraction to be reduced, consequently the pressure drop to be increased and fuel centre temperature increased. Because of the good thermal conductivity of CERMET fuel, the fuel temperature fortunately is not an issue and the fuel temperature is always below the required limits for the considered pin diameter range. The fuel volume fraction can be increased by reducing the pin number per SA and increasing the pin size without significantly changing the overall coolant/clad thermal-hydraulic conditions. An important finding of the study is that the disadvantage of the CERMET fuel in lower "specific" transmutation rate for MA can completely be overcome. Burn-up calculations are performed by applying the TRAIN code [10] with nuclear data JEFF-3.1 to check the reactivity swing and the transmutation efficiency. The thermal-hydraulic calculations have been performed with the SIMMER code [7]. There are several ways to change the pin diameter and the number of pins per SA. One could keep the coolant area, clad thickness and gap unchanged (Option 1). Decreasing the pin number thus increases the hydraulic diameter and reduces the pressure drop. The other variant is to keep the pressure drop unchanged (Option 2) and the clad thickness and gap change proportionally to the change of pin diameter. Both options yield almost the same fuel volume fraction. Therefore neutronic burn-up calculations done for the first option also hold for the second option. For the second option the fuel volume fraction changes from 28.34% to 29.96% and 31.80%, corresponding to a number of pins as 127 (11.274 mm diameter) to 91 (13.695 mm diameter) to 61 (17.233 mm diameter).

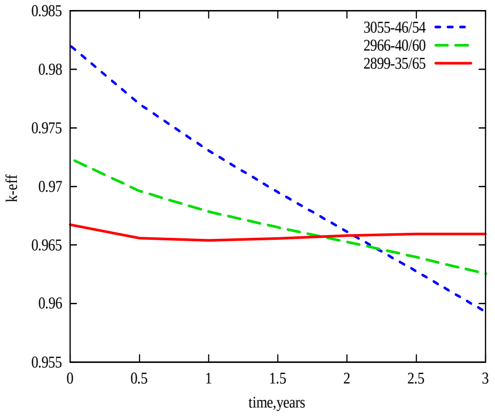
In Figure 3.2 the k_{eff} variation depending on the Pu/MA ratio is displayed. As can be seen in the figure, the reactivity variation over the three years is quite limited, thus the constant beam requirements can be fulfilled.

In conclusion one can state that a Mo CERMET core (shortly CERMET'08 core) can be designed having a very good transmutation performance and minimal reactivity swing with the additional advantage of a matrix with high safety performance. One should note that the uncertainties (in criticality, coolant/structure reactivity effects and burn-up reactivity loss) due to nuclear data remain relatively high. One particular issue relates to the $^{241}Am(n,\gamma)$ reaction which may yield either

Table 3.1: Burn-up calculation results of pin number variation

Pin number per SA	**168+1**	**91/Op 1**	**61/Op 1**	**61/Op 1 + thicker pins in outer zone**
Pu/MA atom ratio overall	**46/54**	**40/60**	**35/65**	**35/65**
Pin diameter (mm)	9.52	11.00	13.00	15.87
Fuel VF in the core and outer zone (%)	26.73	29.79	31.24	31.24/35.75
k_{eff} initial	0.9820	0.9724	0.9428	0.9667
k_{eff} after three years	0.9593	0.9625	0.9455	0.9660
Pu initial mass (kg)	3 055	2 966	2 726	2 899
Ma initial mass (kg)	3 610	4 479	5 056	5 377
Pu consumption [kg/TWhth]	**5.71**	**-1.06**	**-7.95**	**-7.22**
U consumption [kg/TWhth]	-0.48	-0.49	-0.50	-0.51
Am consumption [kg/TWhth]	46.80	54.72	63.40	62.40
Cm consumption [kg/TWhth]	-9.70	-10.70	-12.27	-11.76
Np consumption [kg/TWhth]	0.86	0.96	0.60	0.44
MA consumption [kg/TWhth]	**37.96**	**44.98**	**51.73**	**51.08**

Figure 3.2: K_{eff} variation over three-year burn-up for different Pu/MA ratios



^{242}Am or ^{242m}Am: the branching ratio (the ^{242}Am to ^{242m}Am production ratio) varies from ca. 0.92/0.08 (in models based on JEFF-3.1/EAF activation data files) to ca. 0.85/0.15 (in models adopted at CEA). The branching ratios also influence the reactivity loss per cycle. Thus, particular attention should be paid to the branching ratio of the ^{241}Am capture reaction, as uncertainties in this value influence significantly the reactivity loss due to fuel burn-up [31].

The safety parameters have been calculated with SIMMER-III [7,8]. The void worth is calculated with 7 335 pcm, the β_{eff} with 192 pcm and the Doppler constant is given with -68 pcm. The void worth is usually a bit overestimated by SIMMER using an 11-group FZK cross-section library but is of the same order of magnitude as for the CERCER core.

4 Fuel considerations and safety analyses for the CERMET core

The key decision for recommending the CERMET fuel matrix was based on safety performance. For the MgO matrix both the problem of a drastically reduced thermal conductivity especially in the critical high-temperature domain (T-fuel > 1 500 K) and the problem of a possible dissociation at T > 1 950 K (with the impact of fuel redistribution and recriticality) should lead to the placement of MgO only as a back-up solution [20,21]. The recommendation of AFTRA has to be seen in the light of the limited knowledge on these fuels under irradiation or transient conditions. The safety of these fuels and the cores developed can only be judged after the in-flux of experimental data and inclusion of these data into the analyses. One has to note that a significant aspect of these fuels, its micro- and macro-physical behaviour (*e.g.* mechanisms of gas release, swelling, high T redistribution of Am under transient conditions, in-clad corrosion) is not included in the safety analyses done up to now.

The dynamic behaviour of an EFIT-type ADT is characterised by the lack of a prompt fuel feedback effect (Doppler), and significant positive (delayed) reactivity feedback potentials. The coolant void worth both for the MgO and Mo-92 fuel type cores is approximately in the range of 6 000-7 000 pcm, in case the entire core should be voided of coolant. Traditionally, the whole core void values are given as a safety indicator in relation to *e.g.* the Doppler (critical fast reactor) or the subcriticality (ADS). The clad worth potentials are in the range of 2 000 pcm. These reactivity potentials are larger than the assumed subcriticality margin of 3 000 pcm and their safety impact has to be assessed. An extensive and complete safety analysis is under way for the CERCER reference core within the Domain I of EUROTRANS. For the CERMET core, safety calculations are performed with respect to most critical issues only. For events which are considered to be less important, the conclusions will be made on the basis considering the results obtained for the CERCER option. In addition, already ongoing safety analyses both for CERCER and CERMET cores within the AFTRA Domain 2 identified key issues and differences in the safety behaviour of the two alternative fuel cores. In general, for the CERMET core, the fuel safety margins are considered significantly higher.

Due to the lack of detailed experimental data with regard to transient fuel conditions and the phenomenological uncertainties in the high-temperature range, both fuel and clad limiting temperature maps have been defined [3,27-29] for the different defence-in-depth categories. The specific characteristic of the composite fuels is that the individual component has to be respected for categorisation. As the matrix generally represents the "continuous phase" and is thus the mechanically stabilising structure the disintegration point of the matrix is a key safety criterion. Both the CERCER MgO- and CERMET Mo-based fuels have much lower melting points than MOX fuel. Fuel melting could be real melting, disintegration/evaporation of one individual component and/or eutectic formation. It has been decided that Category IV events [16] should not lead to fuel "melting". Besides the fuel limits, clad limits of EFIT are of major interest. For evaluating the results of the transient calculations, for CERCER the Category IV "fuel" temperature is 1 950 K and the DEC temperature is given with 2 130 K [3,27-29]. A concern of MgO is the identified potential for a fuel matrix destabilisation in the high-temperature range. This could lead to a disintegration of the fuel under a severe transient, to an in-pin rearrangement of the TRU with a subsequent compaction. In the MgO case the matrix material thus tends to disintegrate before the TRU "fuel phase". For the CERMET the Category IV temperature is 2 400 K and DEC is conservatively given with 2 450 K [3,27-29]. Besides the fuel limits, clad limits of EFIT are of major interest. Clad creep induced fuel pin failures for unirradiated T91 are given here for the CERMET core in Table 4.1.

Table 4.1: Creep failure temperature limits for T91 steel for the Mo-92 pin design

Failure time/ gas pressure	0.1 s	1 s	10 s	2 min	30 min
1 MPa	1 342 K	1 279 K	1 222 K	1 166 K	1 111 K
5 MPa	1 315 K	1 253 K	1 198 K	1 143 K	1 088 K

One should note that the creep failure data are derived and extrapolated from a Larson-Miller parameter correlation, which is an approximation, mostly applicable for the non-transient domain. In addition some recent results of experiments under HLM conditions indicate a further reduction [32] of the creep failure temperatures for T91. In conclusion the values given in Table 4.1 can only be regarded as approximate.

For the CERMET core, safety analyses have been performed for various protected and unprotected transients as P/UTOP protected/unprotected transient over power, UTOC (unprotected over current), beam trip (BT) and ULOF (unprotected loss of flow) and UBA (unprotected blockage accident). The UBA and the ULOF are presented here to highlight the differences in possible voiding phenomena caused by pin failures.

4.1 Unprotected blockage accident (UBA)

For the blockage accidents, in case of pin failures, the release of helium, which may cause voiding and a reactivity increase, could be a concern in an ADT, as no limiting Doppler effect is available during a transient which would use up a considerable part of the void worth of the core. The following scenario is predicted in SIMMER-III simulations. The blockage location in the Mo92-CERMET cores is chosen at the core inlet of the first fuel ring and the coolant mass flow rate reduced to 6.5% in the blocked channel. Clad failure causes a gas blowout but the fuel is still covered by the clad and clad removal is assumed to occur at around 1 700 K when the cladding loses its strength. The fuel might then break up into fuel chunks with a particle diameter ranging from 0.5 mm to the pellet size. Can-wall temperatures are also in the range close to melting point. Figure 4.1 shows the power and reactivity trace and Figure 4.2 shows the void development of the transient. Fortunately, the gas release and voiding does not induce a very significant reactivity/power increase, because of the incoherence of voiding and the rewetting after the local gas blow-out.

Figure 4.1: Power and reactivity transient in an UBA (Mo-92 core)

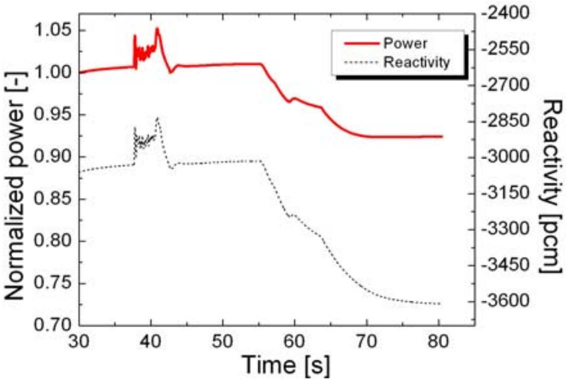
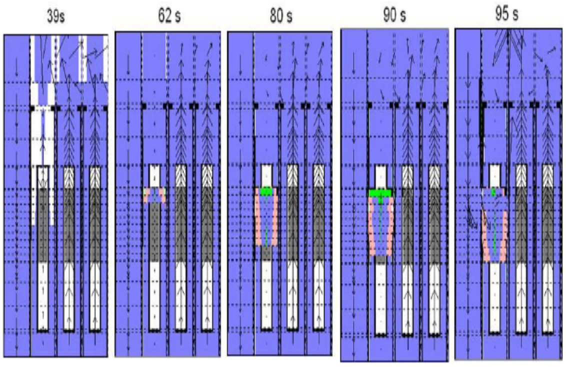


Figure 4.2: Void distribution, rewetting and fuel break-up in an UBA (Mo-92 core)



After clad disruption and fuel pin failure, broken fuel particles can be swept out from the blocked fuel ring reducing the reactivity and power. The decrease of power is limited in a source driven system but helps to allow for a longer grace time to shut down the beam before large-scale damage propagation might commence. The failure temperatures of the cladding (failure temperature for gas blow-down and temperature for release of fuel pellets or chunks from pellet stack after loss of strength), the behaviour of the pellets and the influence of the upper pin and bundle structures decide on the outcome of the transient. If the pellets do not break up into smaller chunks the fuel sweep-out from the blocked channel is not possible. A failure propagation is initiated and the wrapper of the neighbouring fuel assemblies melts. Similar phenomena could happen inside the blocked fuel ring with smaller particles when at low clad failure limits massive particle generation takes place which leads to particle jamming and blockage formation. Generally, one would rather expect a non-break-up of the Mo pellets and thus a failure propagation until larger spaces are opened via hexcan and upper structure melting and the fuel pellets can be removed from the core region. A major point is however that due to the incoherence in clad and coolant temperature levels in the blocked and non-blocked assemblies the void release via clad failure is incoherent, includes rewetting and does not lead to a massive and rapid reactivity increase. Further work must be invested for an improved simulation and clad and fuel behaviour in the high-temperature range should be experimentally understood.

4.2 Unprotected loss of flow (ULOF)

Unprotected loss of flow (ULOF) is another transient of interest to show the safety potential of the design. The liquid lead has good natural convection properties. As the primary coolant pump coasts down, natural convection takes over at a certain reduced mass flow rate. The remaining mass flow rate depends mainly on the pressure drop in the primary loop, the coolant temperature difference between the core outlet and inlet and the height of the heat exchanger (HX). The HX height (from core mid-plane to HX mid-plane) is 3.7 m and the nominal temperature rise through the core is 80 K. Analyses show that the ULOF is survived in the CERMET core and the temperature values of the fuel and clad are below the safety limits. Again, the fuel temperatures are by nearly 1 000 K below any failure limits (see Figures 4.3 and 4.4).

Figure 4.3: Power and reactivity transient in a ULOF (Mo-92 core)

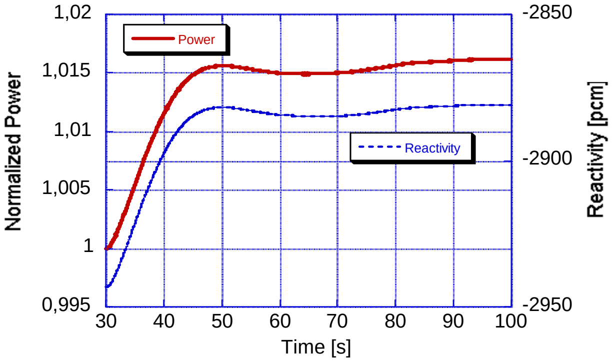
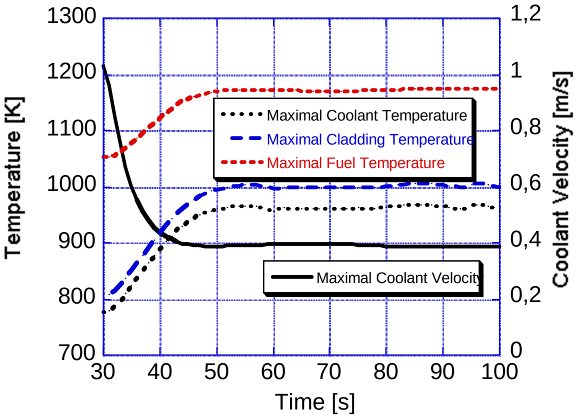


Figure 4.4: Temperature levels for fuel, clad and coolant in a ULOF (Mo-92 core)



As briefly described before, a CERMET core has been investigated doubling the power density. This case was also of interest from the safety point of view, as failure phenomena of clad and the impact on a possible void worth development could be studied. In the calculation shown (Figure 4.5), the plenum pressure has been reduced to 30 bars to reduce the blow-down and voiding potential of fission gas and He after pin failure. Clad failure took place under ULOF conditions in this high-power core and subsequent voiding leads to a power surge with additional pin failures and propagation. In Figure 4.5 the power and reactivity development is given and in Figure 4.6 the growth of the void region under ULOF conditions is displayed. A clear pin failure propagation scenario is identified. The stepwise void worth activation and failure progression can already be identified in the reactivity trace. The main problem is with the low resistance of the T91 clad to high-temperature creep failure.

In all calculations it is not the fuel that poses the limiting conditions for failure, but rather the clad. The difference in the scenario to an UBA should be noted. While under UBA conditions voiding is very incoherent and not capable to drive a significant and rapid failure propagation, in the ULOF the coherency of the temperature levels is significantly larger and rapid failure propagation occurs with reactivity addition. Under UBA conditions after a local gas release, the core is rapidly rewetted by lead, which is different under ULOF conditions. The ULOF thus has an intrinsic potential for triggering a power excursion. The results of these calculations are consistent with earlier findings for a large ADS where the high void worth and its activation lead to a severe power transient in the simulation [34,35].

Figure 4.5: Power and reactivity transient in a ULOF (Mo-92 core with double power)

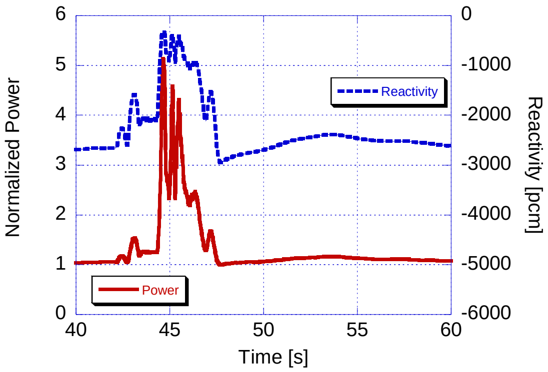
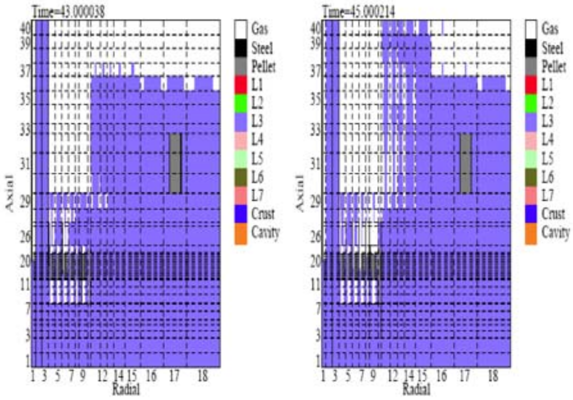


Figure 4.6: Void distribution and development in a ULOF (Mo-92 core with double power)



The conclusions of the present safety investigations can be summarised as follows:

- The CERMET EFIT can survive a ULOF transient.
- In case the EFIT power density is increased (in the analysed cases by a factor 2) the ULOF transient leads to clad failure conditions and a pin failure propagation commences. The fuel itself is far below any temperature limits. In the ULOF, the coherency of the temperature levels is significant and rapid failure propagation occurs with reactivity addition and a power surge. The ULOF thus has an intrinsic potential for triggering a severe power excursion. Other transients with the potential of a coherent coolant and clad temperature increase could reveal a similar failure potential.
- Besides creep failures other failure mechanisms are not taken into account but should be in further analyses.
- The unprotected blockage accident (UBA) is a path into core disruption with limited damage propagation because of incoherency effects. Voiding does not induce a severe power surge even under failure propagation conditions. Fuel sweep-out and a reactivity and power reduction can only be expected if the pellet would break up into small particles after becoming unclad.

5 Conclusions

The EFIT, the European Facility for Industrial Transmutation, developed within the EUROTRANS programme, aims at a generic conceptual design of a full transmuter. The EUROTRANS Domain DM1 (DESIGN) developed the conceptual reference design of the EFIT, a 400 MW_{th} ADT, loaded with a CERCER, U-free fuel based on an MgO matrix with a zero Pu balance (42:0 concept). The design has been worked out in all details and as reference design it is currently subjected to an extensive safety assessment. The Domain DM3 (AFTRA), responsible for the fuel development within EUROTRANS, in parallel developed a core loaded with a Mo-92 CERMET matrix-based fuel. Because the main mission of AFTRA is fuel development, the level of detail in the design work is not comparable with the CERCER core. In both cores, for the cladding, the 9Cr1MoVNb T91 steel has been chosen. The core coolant is pure lead with inlet and outlet temperatures of 673 K and 753 K. The EFIT concept was intended to be optimised towards: good transmutation efficiency, high burn-up, low reactivity swing, low power peaking, adequate subcriticality, reasonable beam requirements and a high level of safety. The two designs are reported and discussed. It is shown that with the CERMET fuel a similarly good transmutation performance of MA with a low reactivity burn-up swing can be achieved when the thermal property advantages of a metal matrix are exploited. The main reason for recommending CERMET by the AFTRA Domain lies in its higher safety potential. For the CERMET core the most important safety analyses have already been performed for BOL core conditions, sufficient for a preliminary safety assessment. In no case were any safety limits for fuel surpassed. Both the CERCER and the CERMET core reveal high void worth potentials, larger than the built-in core subcriticality. The impact of local void activation via pin failures and gas blow-down has been investigated. Important differences in the void release scenarios have been identified for typical accidents to be investigated within the safety assessment, *e.g.* the unprotected loss of flow (ULOF) or the unprotected blockage accident (UBA). In general, the T91 clad, but not the fuels, set the limiting conditions in design flexibility and safety. For the EFIT a corrosion protective GESA-type treatment for the structures is assumed. Further experimental information both on clad and fuel will become available to reduce the uncertainties in the safety assessment. Of major importance is the clad issue where data in the high-temperature domain on T91 are needed and alternative claddings have to be investigated. Within AFTRA especially the in-pile experiment FUTURIX will provide information on the irradiation behaviour of CERCER and CERMET fuels and HELIOS and BODEX will provide information on the impact of the He release during operation and under shutdown conditions. Further work can be envisioned in optimising the EFIT design, *e.g.* improving the source efficiency and/or increasing the power density to some extent. The impact of irradiation and burn-up conditions on safety is another issue to be investigated. Tailoring the EFIT to the needs of different countries (phase-out versus ongoing nuclear development) within global and/or regional scenarios is also of interest.

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Design study of minor-actinide-bearing oxide fuel core for homogeneous TRU recycling fast reactor system

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Abstract

In the Fast Reactor Cycle Technology Development Project (FaCT) in Japan, sodium-cooled fast breeder reactors (FBR) with mixed-oxide fuel were selected as the most promising candidate for commercial power reactors. From the viewpoints of sustainability and waste management, a core design study for the FaCT reactor has been performed in terms of the recycle of transuranium elements (TRU), namely, plutonium and minor actinides (MA: Np, Am, Cm). The TRU recycle mode is so-called "homogeneous" in which all the FBR contain MA-bearing fuel. The homogeneous TRU recycle has the following advantages: there is no prominent influence of MA on core and fuel design, therefore target of required R&D does not become significantly deep. The present study focused on the influence of TRU composition change on core design during the transition stage from light water reactors (LWR) to FBR. There is a possibility for FBR fuel to have degraded Pu and MA recovered from LWR spent fuel. MA contents in the fuel will vary from 1-5% depending on the type of LWR, internal storage period of spent fuel, and the number of FBR recycle times. As a result, it is found that about 30~40% of MA can be transmuted during the residence period in the FaCT reactor if the MA content in fresh fuel is 3~5%. Degraded Pu and MA deteriorate the safety-related reactivity coefficients such as sodium void reactivity. Core design modifications may be necessary to cope with those influences. Fuel design perspectives are also mentioned since MA affects the fuel design by reducing the margin to fuel melting and raising inner gas pressure due to helium generation. Along with FBR recycling, due to the accumulation of source nuclides such as ^{244}Cm, we might have to deal with the influence of decay heat on the fuel fabrication and the transport of fuel subassemblies. The homogeneous TRU recycle has another advantage that it can provide a feasible solution to the decay heat problem.

Introduction

Fast breeder reactors (FBR) can potentially reduce the environmental burden of radioactive waste effectively by the recycle of minor actinides (MA) in spent fuel. In the Fast Reactor Cycle Technology Development Project (FaCT) in Japan, the conceptual design studies of FBR cycle system have been performed in terms of the recycle of transuranium elements (TRU), namely, plutonium and MA (Np, Am, Cm), from the viewpoints of sustainability and waste management [1]. The TRU recycle mode is so-called "homogeneous" in which all the FBR contain MA-bearing fuel.

In the FaCT project, the Japanese sodium-cooled fast reactor (JSFR) with mixed-oxide (MOX) fuel was selected as the most promising candidate for commercial power reactors. The reference MOX fuel core is a "high internal conversion" type core [2]. This advanced concept has an economical advantage by achieving high total average discharge burn-up (including blanket) and sufficient breeding ratio with small amount of blanket. Design studies of the reference core had been mainly performed for the multi-recycle equilibrium stage of FBR.

The current conceptual design study focuses on the influence of TRU composition change on FBR core design during the transition stage from light water reactors (LWR) to FBR. In homogeneous recycling, MA contents in FBR fuel will vary from 1-5 wt.% (per cent in heavy metal weight) depending on LWR type, internal storage period of spent fuel, and the number of FBR recycle times. Design study for MA-bearing fuel core has so far been performed assuming 3 wt.% of MA content in fresh fuel as a typical case [3]. The results are presented in this paper. Design issues and perspectives are discussed for the case of higher MA content that would be expected in the transition stage.

Finally, some remarks are made about the merits and demerits of homogeneous recycling, in comparison with heterogeneous recycling that employs MA target assemblies or dedicated reactors.

Design conditions for FaCT reactor cores

In the beginning of the FaCT project, the development targets and requirements were reviewed and re-established for both commercialised fast reactors and fuel cycle facilities [4]. Design conditions, targets, requirements and limits of the present core design study are summarised in Table 1.

Plant conditions

In this study, a large-scale reactor of 3 530 MWt is mainly evaluated. The core outlet and inlet temperatures are 550°C and 395°C, respectively. The envelope diameter of radial shielding is about 7.0 m or less.

Core performance targets

Flexible breeding ratios from 1.1 to 1.0 are targeted, which is needed for smooth transition from LWR to FBR and for sustainability after the transition. Though a high breeding core whose breeding ratio is 1.2 is also required in the FaCT project, it is out of scope in the present study. The target of total average discharge burn-up (including blanket) is more than 80 GWd/t. High total average discharge burn-up contributes to the reduction of fuel cycle cost.

Safety requirement

The MOX fuel core design aims for sodium void reactivity about 6$ or less, core height 100 cm or less, and average core specific heat about 40 kW/kg-MOX or more. These are preliminary target values to prevent the super-prompt criticality in the initiating phase of core disruptive accident (CDA) based on the experience of sodium-cooled reactor core safety evaluation [5]. In addition, the Fuel Assembly with Inner Duct Structure (FAIDUS) fuel subassembly is adopted as a measure of early discharge of molten fuel to prevent re-criticality in the transition phase of CDA.

Table 1: Major design conditions for large-scale MOX fuel cores in FaCT Project

Item	Value
Plant conditions	
Power output [MW_e/MW_t]	1 500/3 530
Coolant temp. [°C] (outlet/inlet)	550/395
Shielding region diameter [m]	< about 7.0
Core performance targets	
Breeding ratio	1.1~1.0
Discharge burn-up [GWd/t]	
Core	150
Total (including blanket)	> 80
Operation cycle length [month]	> 24
Safety requirements	
Sodium void reactivity [$]	< about 6
Core height [cm]	< 100
Average core specific heat [kW/kg-MOX]	> about 40
Subassembly concept for re-criticality free	FAIDUS
Fuel specifications	
Fuel composition	Low-decontaminated TRU fuel with residual FP
Fuel smear density [%TD]	82
Cladding material	ODS steel
Thermal hydraulic condition	
Maximum cladding mid-wall temperature [°C]	< 700
Bundle pressure drop [MPa]	< about 0.2
Fuel integrity limits	
Maximum linear heat rate [W/cm]	< about 430
CDF (steady state)	< 0.5
Maximum fast neutron fluence ($E_n > 0.1$ MeV) [n/cm^2]	< about 5×10^{23}

Fuel specifications

Low-decontaminated TRU fuel is applied from the economical and non-proliferation viewpoints. The fuel contains a certain amount of MA which shall be from 1 to approximately 5 wt.% for uranium resource utilisation and environmental burden reduction. The TRU composition will change during the transition stage from LWR to FBR. Two representative compositions were selected for core design study; these are FBR multi-recycle composition and LWR spent fuel composition to be explained in the next section. Residual fission products (FP) are contained in fresh fuel corresponding to the decontamination factor for an applied reprocessing system. The content of residual FP in the FBR multi-recycle stage was estimated as 0.2% in volume for MOX fuel. The conent for the transition stage depends on the recycle condition of LWR spent fuel.

Oxide-dispersion Strengthened (ODS) steel is selected as fuel cladding material, because it can withstand high neutron fluence and has excellent high-temperature strength [6,7].

Thermal-hydraulic condition

The maximum cladding mid-wall temperature of MOX fuel core is limited to 700°C or less in accordance with the development target of ODS steel. Fuel pin bundle pressure drop is as low as about 0.2 MPa, which facilitates the natural circulation capability of the plant primary coolant system.

Fuel integrity limits

The design limits of linear heat rate are about 430 W/cm for MOX fuel core. The fuel cladding cumulative damage fraction (CDF) in steady-state operation is 0.5 or less. The current target of fast neutron fluence ($E_n > 0.1$ MeV) of ODS steel is about 5×10^{23} n/cm^2.

The reference core in the FaCT project

The reference MOX fuel core, the high internal conversion (HIC) type core, was designed to achieve low fuel cycle cost and reasonable breeding capability [2]. That is a concept aiming to reduce fuel cycle cost by increasing total average discharge burn-up. As an actual measure, large diameter fuel pins (10.4 mm) are applied to increase the internal conversion ratio and to reduce the amount of blanket as much as possible. Moreover, the HIC-type core enables to extend the operation cycle length due to its high internal conversion ratio, which is also an economical advantage. Major core and fuel specifications are summarised in Table 3.

As for breeding capability, a breeding ratio of 1.1 can be obtained with axial blanket and only one layer of radial blanket subassemblies. It is possible to alter the breading ratio to 1.03 by replacing the radial blanket with steel shielding and by adjusting the axial blanket thickness. The core configurations are illustrated in Figure 1. The HIC-type core can flexibly comply with the changes of plutonium breeding requirement.

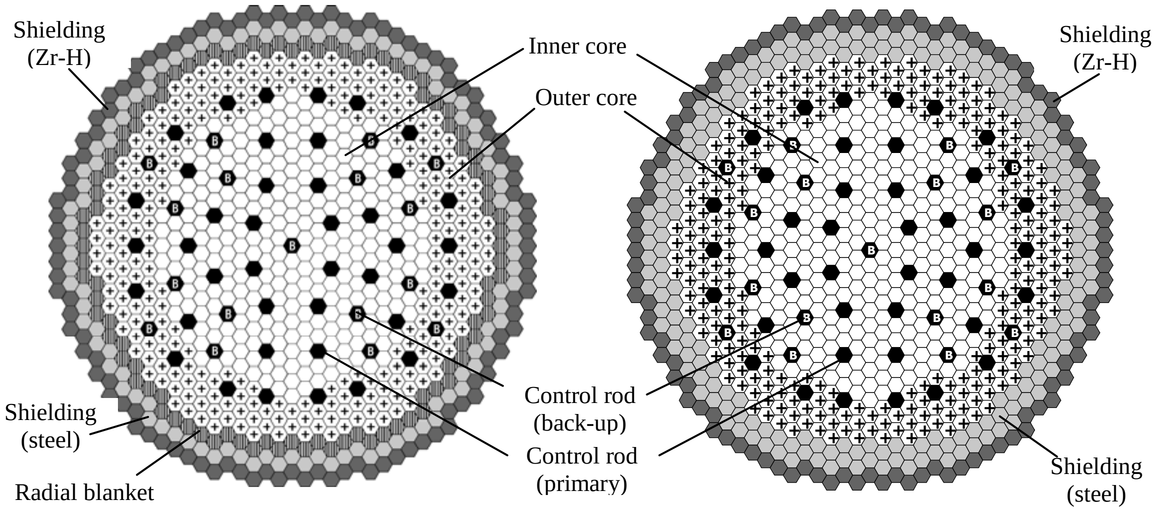
The HIC-type core adopts an innovative fuel subassembly called FAIDUS as a measure of early discharge of molten fuel to prevent re-criticality in the transition phase of CDA [8]. As illustrated in Figure 2, an inner duct is installed at a corner of the subassembly, and a part of the upper shield is removed. Molten fuel enters the inner duct channel and goes out into outside through the upper shield. The FAIDUS-type subassembly is expected to have superior performance for the discharge of molten fuel in terms of preventing compaction.

Design study for MA-bearing fuel core

A design study of MA-bearing fuel core was carried out based on the reference HIC-type core. As for a representative TRU composition, we chose the composition recycled from LWR spent fuel (ALWR, average burn-up of 60 GWd/t) cooled over 40 years, which is listed in Table 2. It is the most influential composition on the safety-related reactivity coefficients in a standard transition scenario from LWR to FBR, providing that the maximum MA content in fresh fuel is 3 wt.%.

In this study, Am and Cm were assumed to be recovered separately by an MA extraction process, and Np was assumed to be co-recovered with Pu. Am-Cm and U-Pu-Np were blended so that the total MA content would be 3 wt.%. This MA recycling method is one of the options considered in the FaCT project. The residual FP content was tentatively assumed to be 0.6% in fuel volume.

Figure 1: Core configurations of the large-scale "high internal conversion" type MOX fuel core



[Breeding ratio: 1.1]

[Breeding ratio: 1.03]

Figure 2: Conceptual view of FAIDUS-type fuel subassembly

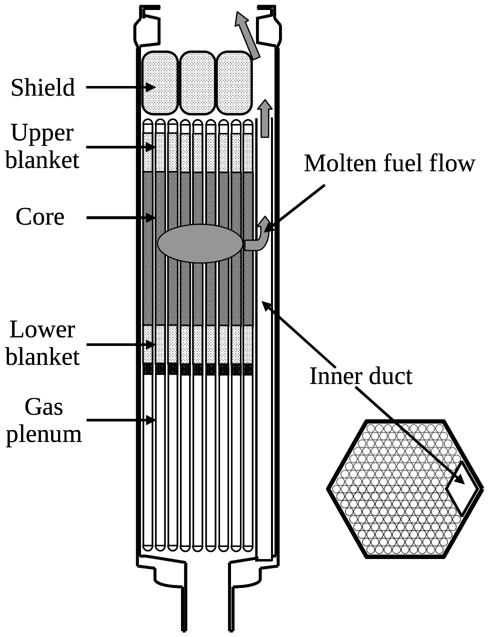


Table 2: TRU compositions for core design study (in weight per cent)

Nuclide	FBR multi-recycle composition	LWR spent fuel composition
^{238}Pu	1.7	2.8
^{239}Pu	55.9	50.0
^{240}Pu	30.5	27.3
^{241}Pu	3.4	1.8
^{242}Pu	3.3	9.2
^{239}Np	0.3	8.9
Subtotal	–	100.0
^{241}Am	3.0	83.4
^{242m}Am	0.1	0.1
^{243}Am	1.0	14.9
^{244}Cm	0.6	1.2
^{245}Cm	0.2	0.4
Subtotal	–	100.0
Total	100.0	–

Core characteristics have been calculated using the neutron diffusion theory for three-dimensional Tri-Z geometry. Self-shielded cross-sections were prepared for a homogeneous cell representation by using an adjusted cross-section library ADJ2000R [9,10]. To evaluate design values, the following corrections were taken into account: neutron transport correction, spatial mesh correction, cell heterogeneity correction, as well as design margins. Integral experimental information of critical facilities and existing reactors are already fed back into ADJ2000R so that we can obtain the best-estimated design values without any bias factor corrections.

The main core specifications and characteristics of the MA-bearing fuel core are shown in Table 3, in comparison with those of the reference core that uses the multi-recycle TRU composition (cf. Table 2) [3,11]. Since MA as well as fertile Pu isotopes improve the burn-up characteristics, the upper axial blanket was eliminated by 5 cm to yield the required breeding ratio 1.1. On the other hand, they deteriorate the safety-related reactivity coefficients; the sodium void reactivity increases to 5.9$, which still satisfies the safety requirement.

Table 3: Major specifications and characteristics of MA-bearing fuel core

Item	Reference core	MA-bearing fuel core
Power output [MW_e/MW_t]	1 500/3 530	←
Core height [cm]	100	←
Axial blanket thickness [cm] (upper/lower)	20/20	15/20
Number of fuel subassemblies (inner core/outer core/ radial blanket)	288/274/96	←
Operation cycle length [month]	26.3	←
Fuel exchange batch (core/radial blanket)	4/4	←
Fuel pin diameter [mm]	10.4	←
Number of fuel pins per subassembly	255	←
Gas plenum length [mm] (upper/lower)	100/1 100	100/1 150
TRU composition (see Table 2)	FBR multi-recycle	LWR spent fuel
Pu enrichment [wt.%] (inner core/outer core)	18.2/20.6	19.6/22.1
MA content [wt.%] (inner core/outer core)	1.0/1.1	3.0/3.0
Pu fissile inventory [t/GW_e]	5.8	5.7
Burn-up reactivity [%Δk/kk′]	2.5	1.8
Breeding ratio (total)	1.1	1.1
(core/axial blanket/radial blanket)	0.83/0.19/0.07	0.84/0.19/0.07
Average discharge burn-up [GWd/t] (core/total)	145/89	146/93
MA transmutation amount per batch [kg]	21	205
MA transmutation rate [%]	11	38
Average core-specific heat [kW/kg-MOX]	40	40
Sodium void reactivity (EOEC) [$]	5.2	5.9
Doppler coefficient (EOEC) [10^{-3} Tdk/dT]	-5.7	-4.5
Maximum linear heat rate [W/cm] (inner core/outer core)	403/402	411/395
Maximum fast neutron fluence* [n/cm^2]	4.9×10^{23}	5.0×10^{23}
Maximum cladding mid-wall temperature [°C]	700	700
Maximum bundle pressure drop [MPa]	0.22	0.22
Maximum CDF value	0.49	0.45

* E_n > 0.1 MeV, two-dimensional calculation.

MA transmutation amount and rate are defined as follows:

- MA transmutation amount [kg]: $M_i - M_f$;
- MA transmutation rate [%]: $(M_i - M_f)/M_i$.

M_i and M_f stand for MA-loading and discharged amounts per one fuel exchange batch, respectively. For the present MA-bearing fuel core, the MA-loading amount per one fuel exchange batch is 537 kg. The MA transmutation amount is 205 kg. Then, the MA transmutation rate turns out to be 38%.

High Am (and Cm) content may affect the fuel designs; the limit of linear heat rate is reduced due to the decreases of melting point and thermal conductivity of fuel; the gas plenum length may have to be extended due to the increase of helium gas generation from ^{241}Am capture reaction and subsequent decay process. Naganuma, *et al.* [12,13] reflected the recent experimental data evaluations for melting point and thermal conductivity of Am-containing MOX fuel to the evaluation of linear heat limitations. As a result, it was found that the decrease of the limit of linear heat rate is small (several W/cm from the reference core) and the maximum linear heat rate satisfies the limitation. Moreover, it was found that a 50 mm extension of gas plenum length is sufficient to satisfy the CDF limitation for the present MA content and the recycling method of LWR spent fuel [12,13]. The increase in gas plenum length and the elimination of the axial blanket cancel each other out, thus the total fuel pin length is maintained.

In this way, it is confirmed that the HIC-type core enables to accept the typical MA-containing fuel with slight modifications to the core specification. There is no prominent influence of MA on core and fuel design and then the R&D target does not become significantly deep, which are part of the advantage of homogeneous recycle.

In the future, we will conduct a core design study with higher MA content (more than 3 wt.%). The present design result suggests that the sodium void reactivity and the fuel cladding CDF would be the key points.

TRU mass balance calculation in a homogeneous recycle scenario

TRU mass balance and its composition change during the LWR-FBR transition phase have been evaluated by homogeneous-recycle scenario calculations [14]. By using a typical case from those calculation results as an example, MA transmutation rate and the decay heat of fresh fuel were evaluated for the reference core.

In a typical homogeneous recycling scenario, domestic nuclear power generation capacity was assumed to be 58 GW_e. LWR were decommissioned after 40 to 60 years operation and ALWR were introduced after 2030. The plutonium recycling in LWR ended at around 2045. After 2050, LWR of about 1 GW_e was replaced by FBR every year, and the replacement to FBR was almost completed at the beginning of the 22nd century.

Changes of MA content and Pu isotopic composition in FBR core fuels during the period from 2050 (the start of FBR deployment) to 2200 are presented in Figures 3 and 4, respectively. Along with the FBR recycle, the MA content decreases due to the transmutation of Np and Am. We can recognise an accumulation of small content of Cm. Plutonium seems to increase its fissile component because of the mixture of higher-grade Pu generated in blanket fuels.

Figure 3: MA content change after the start of FBR deployment (an example)

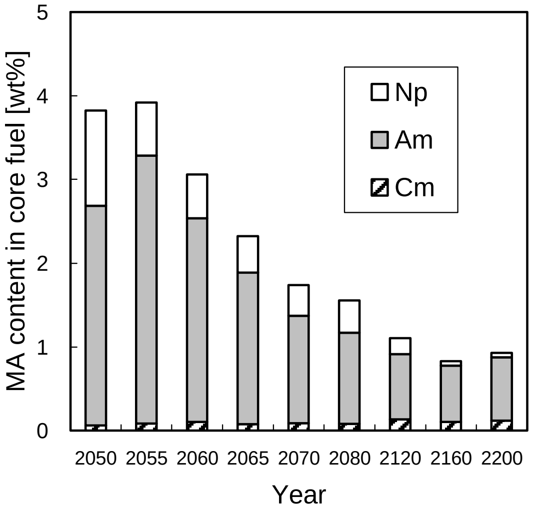
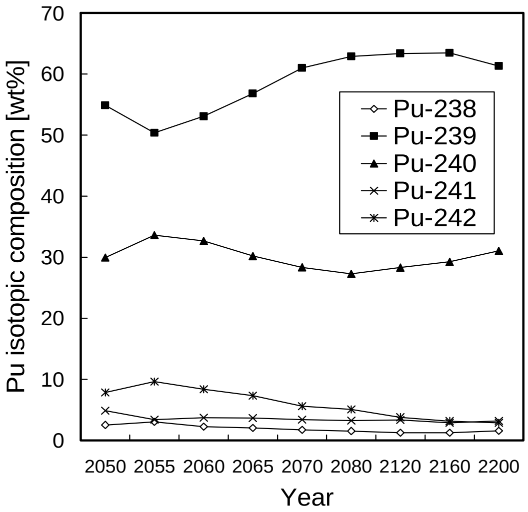
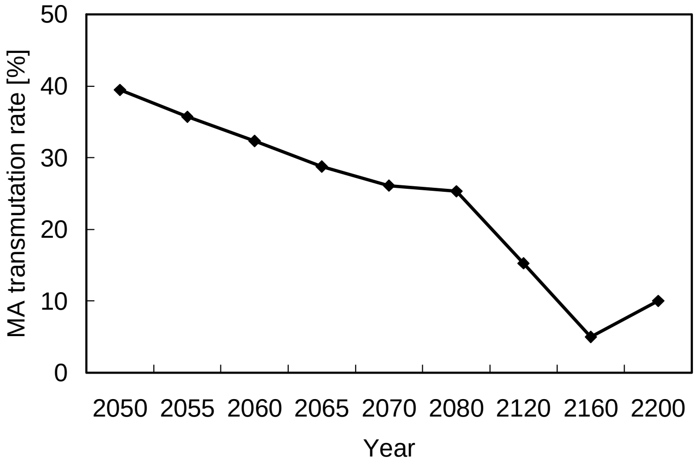


Figure 4: Pu isotopic composition change after the start of FBR deployment (an example)



The change in the MA transmutation rate during that period is shown in Figure 5. It is found that about 30~40% of MA can be transmuted in the reference core if the MA content in fresh fuel is 3~5%. The transmuted MA amount corresponds to the MA from 3~4 LWR of the same reactor power. The MA transmutation rate tends to decrease along with recycling and approaches the level of the FBR equilibrium stage.

Figure 5: Change of MA transmutation rate after the start of FBR deployment (an example)

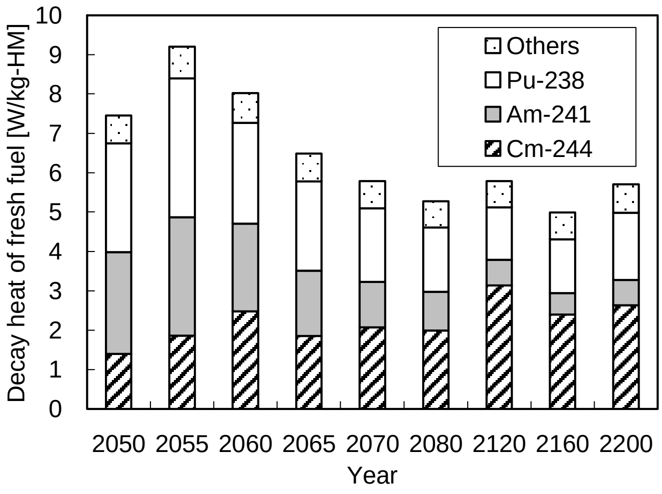


As the content of fertile nuclides such as ^{237}Np and ^{241}Am tend to decrease year by year, the sodium void reactivity is also expected to decrease. Therefore, it is reasonable to select a design point at the beginning of LWR-FBR transition phase for conservative core design in terms of the sodium void reactivity.

Since Am (and Cm) as well as ^{241}Am content decrease along with recycling, the beginning of the LWR-FBR transition phase is suitable for the conservative fuel integrity evaluation.

Calculation results of fresh fuel decay heat are presented in Figure 6. ^{238}Pu, ^{241}Am and ^{244}Cm mainly contribute. It should be noted that ^{244}Cm builds up along with recycling. Increment of decay heat might bring significant influence on the fuel fabrication and the transport of fuel subassemblies. The present calculation results have enough allowance comparing with the tentative upper limit in the FaCT project, 20 W/kg-HM [15]. If the recycle system is designed not to concentrate those source nuclides on particular fuel, the actinide management could be more feasible.

Figure 6: Change of fresh fuel decay heat after the start of FBR deployment (an example)



Concluding remarks

In the FaCT project, conceptual design studies for a sodium-cooled MOX fuel core for JSFR have proceeded with a focus on the TRU composition change during the transition stage from LWR to FBR, where so-called homogeneous TRU recycling is selected. It has been confirmed that the reference "high internal conversion" type core enables to accept the typical MA-containing fuel (up to 3 wt.% of MA content in heavy metal) with slight modifications to the core specification. Moreover, the core has an MA transmutation potential such that about 30~40% of MA can be transmuted during the residence period if the MA content in fresh fuel is 3~5%.

Some of the advantages of homogeneous TRU recycle have been pointed out, in particular:

- there is no prominent influence of MA on core and fuel design;
- target of required R&D does not become significantly deep;
- it has capability of a solution to the decay heat problem in fuel fabrication and handling.

Nevertheless, the heterogeneous recycling that restricts MA treatment to small-scale fuel cycle system might have rationality and one can possibly expect cost benefits. Additional comparative study between homogenous and heterogeneous recycling is necessary before a decisive conclusion can be formed.

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Core performances and safety implications of TRU burning: Medium to large fast reactor core concepts

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Abstract

To investigate the performance parameters and safety implications of reactivity coefficients in large monolithic sodium-cooled fast reactors for a TRU burning, cores whose powers range from 600 MWe to 1 800 MWe were designed. For a TRU burner design, the major design variables to be used in changing the conversion ratio of a given core design are a variation of the core height and fuel cladding thickness. Three power levels of 600, 1 200 and 1 800 MWe were selected to investigate the dependency of the core performance parameters and the reactivity coefficients on the power level. For a consistent comparison, the active core height was adjusted to make the sodium void worth lower than 7.5$, and the number of assemblies was adjusted to attain a similar linear power of around 180 W/cm. The calculation results show that the consumption rate is increased at almost the same rate as the increased power so it seems to have no preference at any power level with the same TRU enrichment. The reactivity coefficients also have little influence on the power level as long as the sodium void worth and the TRU enrichment were calculated to remain to the same value.

Introduction

A Korea Atomic Energy Research Institute (KAERI) study on the Passive Decay Heat Removal Circuit System (PDRC), which is patent pending, showed that there is no virtual limit for the core power from the standpoint of a passive decay heat removal. Thus, the KAERI team will endeavour to achieve a core power level as high as possible without penalising a core safety. With the accumulated design data, accompanied by a safety analysis, the results of this task will be able to identify the most limiting factor in scaling up burner core concepts.

For this purpose, the conceptual core designs of large monolithic sodium-cooled fast reactors for a TRU burning with a power range from 600 to 1 800 MWe have been established. Thereby, it would be possible to provide guidance to future R&D directions for an economic burning of TRU and to achieve maximum benefits from the viewpoint of a core size.

KAERI has been developing the KALIMER-600 [1] core design with a break-even fissile conversion ratio. The core is loaded with a ternary metallic fuel (TRU-U-10Zr), and the break-even characteristics are achieved without any blanket assembly. This core produces a rated thermal power of 1 500 MWt. The KALIMER-600 conceptual design adopts a single fuel enrichment as an alternative to the traditional enrichment zoning approach to flatten a power distribution. As a means to flatten the power distribution, fuel pin designs of different cladding thicknesses are used for different core regions, with the same cladding outer diameter. The use of a single enrichment yields a rather uniform conversion ratio throughout a core. As a result, the power distribution is almost constant over a cycle. The cycle-by-cycle power distribution is also invariant since the charged and discharged fuel assemblies have almost identical worths.

The rather constant power distribution over a cycle makes it possible to achieve a uniform coolant outlet temperature distribution through an appropriate flow allocation to individual assemblies. Consequently, the average coolant outlet temperature can be increased without violating the thermal design criteria, which in turn increases the thermal efficiency. A drawback of this single enrichment design approach with a variable cladding thickness is a higher fissile loading per thermal output relative to the enrichment splitting approach. Since the plutonium fraction required for a breakeven core is rather constant, this results in a larger active core volume.

For a TRU burner core design, the design approach of a variable cladding thickness was investigated. Core designs used the design constraints related to the current technology database imposed for a TRU enrichment limit (30.0 w/o) and a fast neutron irradiation limit (4.0×10^{23} n/cm^2). The benefit of using a single enrichment in this design concept is that it helps to maintain the charged TRU enrichment lower than the case for an enrichment splitting. Therefore, on average a TRU burning can be maximised within the TRU enrichment limit.

For the core design concepts, three power levels of 600, 1 200 and 1 800 MWe were selected to investigate the dependency of the core performance parameters and the reactivity coefficients on the power level. In the next section, the computational method is described, and it also provides descriptions of several design features and the details of the core performance analysis and the reactivity coefficients. Finally, the conclusion is provided.

Computational methods

All the nuclear designs and evaluations were performed with the nuclear calculation module packages in the K-CORE system which is an integrated modular program. Global reactivity feedbacks were calculated using a series of neutron flux solution calculations for a hexagonal-z geometry. The depletion analysis is done with the equilibrium model of the REBUS-3 code system [2] where the DIF3D [3] module solves the neutron diffusion equation with the HEX-Z nodal method and a 25-group cross-section set to obtain the neutron flux and power distributions. All the nuclear cross-sections are based on a 150-group cross-section library called KAFAX-E66 [4] that was prepared based on the ENDF/B-VI data file using NJOY. The 150-group cross-section library includes the infinite dilute cross-sections and Bondarenko self-shielding factors. Using this cross-section library, the effective multi-group cross-sections for a core analysis are produced using the TRANSX [5] code where a self-shielding effect is treated by the Bondarenko method. All of the reactivity coefficients and the control rod worths are calculated using the 25-group DIF3D (HEX-Z nodal) calculations. In the depletion analysis, the active core region is divided into five axial zones. For each axial zone of each assembly, the isotopic number densities are assumed to be spatially constant. The TRU content in the fuel is determined using the REBUS-3 search techniques such that the multiplication factor is to be 1.002 at an EOEC. The reprocessing strategy assumed a 0.1% loss of TRU and a 100% removal of the fission products except for a 5% recovery of the rare-earth (RE) fission products. The reprocessed fuel materials are sent to the fuel fabrication facility where they are mixed with the external feed materials.

Core design and performance analysis

Description of the core design

The 1 500 MWt (600 MWe) TRU burner core design originates from the KALIMER-600 design which is a break-even core with a single enrichment fuel. In the break-even core design, the purpose of varying the cladding thickness was to flatten the power distribution. For a burner core design, in order to reduce the conversion ratio through an increased enrichment, the cladding thickness was increased to decrease the fuel volume fraction. Because the burn-up reactivity swing is considerably larger in a burner core than a break-even core, the number of control assemblies was increased from 16 to 31. In this study, the peak linear power limit was conservatively set at a value of 350 W/cm that yields a cladding inner wall temperature of 650°C for a 30 w/o TRU enrichment fuel.

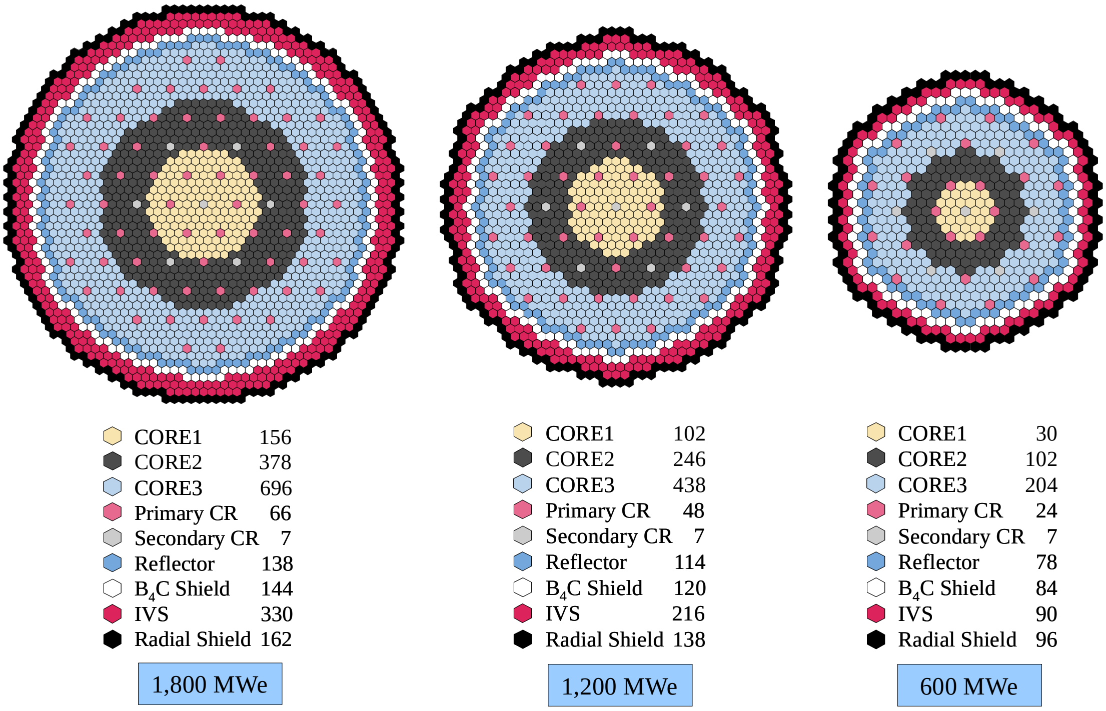
Key design parameters are displayed in Table 1. The Active core height was adjusted to make the sodium void worth lower than 7.5$ to be 85.0 cm, 73.5 cm and 70.0 cm for the 600, 1 200 and 1 800 MWe cores respectively. A cladding outer diameter of 9.0 mm for the breakeven core is reduced to 7.0 mm. The cladding thicknesses are adjusted to make the TRU enrichment as close to 30 w/o as far as possible to promote a TRU burning for the inner (CORE1), middle (CORE2) and outer (CORE3) core regions, respectively. With these adjusted cladding thicknesses, the peak-to-average power ratio that occurs at a BOEC was determined to be 1.52, yielding a peak linear power density of 314 W/cm for the 600 MWe core.

Table 1: Core performances

Design parameter	600 MWe	1 200 MWe	1 800 MWe
Core thermal power (MWt)	1 500	3 000	4 500
Coolant temperature (°C) – inlet/outlet	390/545	390/545	390/545
Number of fuel assemblies	336	786	1 230
Assembly pitch (cm)	16.1	15.9	15.9
Fuel outer diameter (mm)	7.0	7.0	7.0
Pin pitch (mm)	8.890	8.792	8.792
P/D ratio	1.270	1.256	1.256
Cladding thickness (mm) – inner/middle/outer	1.05/0.91/0.77	1.05/0.91/0.77	1.05/0.91/0.77
Eq. core diameter (m)	3.09	4.68	5.86
Eq. reactor diameter (m)	4.51	6.31	7.61
Charged TRU (w/o)	29.92	29.16	28.92
Conversion ratio (fissile/TRU)	0.74/0.57	0.76/0.58	0.76/0.59
Burn-up reactivity swing (pcm)	3 671	3 512	3 508
Cycle length (EFPD)	332	332	332
Sodium void worth (BOEC/EOEC)	6.68/7.28	6.91/7.52	6.87/7.55
Peak fast neutron fluence (n/cm^2)	4.64	4.31	4.42
Max. pressure drop (MPa)	0.156	0.136	0.134
Max. cladding inner wall temp. (°C)	591.27	576.02	571.73
Average linear power (W/cm)	180.4	178.1	179.1
Power peaking factor	1.516	1.476	1.545
Active core height (cm)	85.0	73.5	70.0
TRU consumption rate (kg/cycle)	201.4	384.9	569.5

Higher-power designs were developed by maintaining an average linear power at 180 W/cm in the 600 MWe core. Using the fuel rod design of each region determined in the 600 MWe design, the number of assemblies in each region was determined to minimise the peak-to-average power ratio in the core. The total number of control assemblies was determined to maintain the ratio of the fuel assembly area to the control rod area, with the assumption that the burn-up reactivity swing in the higher-power cores would be similar to that of the 600 MWe core. The numbers of fuel assemblies of the 600, 1 200 and 1 800 MWe designs are 336, 786 and 1 230, respectively. The respective numbers of control assemblies are 31, 55 and 73. Figure 1 shows this radial core configuration.

Figure 1: Core layout



In the core designs with a variable cladding thickness to reduce the fuel volume fraction, the cladding thickness was increased with a fixed fuel pin outer diameter. This resulted in unnecessarily thick claddings, and the structure volume fraction became very high. In the pancake core design, the fuel volume fraction was varied by changing the cladding thickness. Additionally, the core height was reduced to increase the enrichment further, to promote a TRU burning. A reduction of the fuel volume fraction was used as the primary design variable to increase the enrichment, and a reduction of the core height was used as a secondary means, since the latter increases the burn-up reactivity swing more than the former. In other words, we tried to minimise the degree of a core height reduction within the feasible design domain. By minimising the degree of a core height reduction, the effective core radius can be made smaller, which is beneficial from the view point of a construction cost.

The cladding thickness of each core region and the core height were adjusted to meet the design limits for TRU enrichment and a peak linear power. The minimum fuel diameter is 0.49 cm, when the cladding thickness is 0.105 cm for the inner core region fuel rods. As a means to flatten the power distribution, fuel pin designs of different cladding thickness were used in different core regions, while the same cladding outer diameter was adopted throughout the core.

For a consistent comparison, the active core height was adjusted to make the sodium void worth lower than 7.5$, and the number of assemblies was adjusted to attain a similar linear power of around 180 W/cm. The fuel pin diameter and the number of fuel pins were fixed at 7 mm and 271, respectively. However, the pitch to diameter of the fuel rods is allowed to vary. In order to demonstrate a high TRU consumption rate, a core design was evaluated to attain a TRU enrichment of 30 w/o. The major design variables to be used in changing the conversion ratio of a given core design are a variation of the core height and fuel cladding thickness. The cladding thickness was increased to decrease the fuel volume fraction. Simultaneously, the core design was confirmed to have maximum inner cladding temperatures below 650°C and maximum pressure drops below 0.15 MPa. Two independent safety-grade reactivity control systems were employed: primary and secondary control rod systems.

Core performance analysis results

Burning characteristics and performance parameters are listed in Table 1 for the three cores. The REBUS-3 [2] equilibrium model with a 25-group cross-section was used to perform the core depletion analysis.

The calculation results show that large monolithic sodium-cooled fast reactors for a TRU burning, whose power range is from 600 MWe to 1 800 MWe can successfully be designed while meeting all design constraints. The calculated TRU conversion ratio is 0.57 for the 600 MWe core, 0.58 for the 1 200 MWe core and 0.59 for the 1 800 MWe core. The 600 MWe core requires a charged TRU enrichment of 29.9% and yields a reactivity swing of 3 671 pcm over a 332 EFPD. The charged TRU enrichment is 29.2% for the 1 200 MWe core and 28.9% for the 1 800 MWe core.

The core designs have almost the same TRU burning rate per power and a burn-up reactivity swing of ~3 500 pcm. The reactivity swing is 3 512 pcm for the 1 200 MWe core and 3 508 pcm for the 1 800 MWe core. The TRU consumption rate is 201 kg/cycle for the 600 MWe core, 385 kg/cycle for the 1 200 MWe core, and 570 kg/cycle for the 1 800 MWe. Thus the consumption rate is increased by 1.9 times for the 1 200 MWe core and by 2.8 times for the 1 800 MWe core, compared to the 600 MWe core. However, these differences are so small that it is judged that the consumption rate per power is invariant. In all three designs, the sodium void worth upon a core plus fission gas plenum voiding turned out to be less than 7.5$.

Core dimensions were also checked from the viewpoint of a construction cost because if the effective core radius can be made smaller, it is beneficial from the viewpoint of a construction cost. A core with a higher power has a disadvantage from an increased effective core radius to conserve the same linear power due to a core height reduction for a reduction of a sodium void reactivity. The active core height was adjusted to make the sodium void worth to be around 7.5$ because of the reduced neutron leakage effect upon a core power increase. Compared to the 600 MWe core, the active core height is decreased from 85.0 cm to 73.5 cm for the 1 200 MWe core and to 70.0 cm for the 1 800 MWe core. On the other hand, the active core diameter is increased from 3.09 m to 4.68 m for the 1 200 MWe core and to 5.86 m for the 1 800 MWe core. Thus, the radial core area is increased by 2.3 times for the 1 200 MWe core and by 3.6 times for the 1 800 MWe core compared to the 600 MWe core. Thus, it seems that there is a disadvantage from the viewpoint of a core area.

For three cores with a power rating, the peak fast fluence is beyond the assumed limit of HT9 cladding. Additional work may be needed to decrease the peak fast fluence such as by a reduced cycle length or average linear power. However, a detailed study to decrease the peak fast fluence was not pursued at this stage.

Reactivity coefficients

Global reactivity feedback resulting from the Doppler effect, uniform radial expansion and sodium voidings in the equilibrium core are given in Table 2. The reactivity coefficients are estimated at a BOEC and an EOEC for the three cores. From this table, the following are observed. The core with an increased power rating has almost the same Doppler coefficient, a less negative axial expansion coefficient, a less negative control rod worth per rod and a more negative radial expansion coefficient.

Table 2: Reactivity coefficients

	600 MWe		1 200 MWe		1 800 MWe	
	BOEC	**EOEC**	**BOEC**	**EOEC**	**BOEC**	**EOEC**
Doppler coefficient [pcm/°C]	$-804.5\ T^{-1.113}$	$-801.6\ T^{-1.109}$	$-819.3\ T^{-1.109}$	$-816.6\ T^{-1.106}$	$-835.1\ T^{-1.110}$	$-834.3\ T^{-1.107}$
Axial expansion coefficient [pcm/°C]	-0.160	-0.170	-0.121	-0.127	-0.109	-0.114
Radial expansion coefficient [pcm/°C]	-0.707	-0.743	-0.735	-0.771	-0.744	-0.780
Sodium density coefficient [pcm/°C]	0.692	0.750	0.702	0.761	0.697	0.761
Control rod worth [pcm/control rod]	-341	-358	-173	-181	-124	-130
Sodium void worth [$]	6.68	7.28	6.91	7.52	6.87	7.55

The same Doppler coefficient with an increased power rating can be explained by the fact that it has the same TRU enrichment because the Doppler effect is mainly attributed to the fertile isotope. As the power rating increases, the core size increases and accordingly, the neutron leakage rate decreases. The reduced leakage rate increases the chance for fission, so it reduces the TRU enrichment necessary for a criticality, but in this case the TRU enrichment is fixed at a maximum enrichment, 30 w/o. Instead of reducing the enrichment, the core height was reduced to compensate for a reduced neutron leakage to increase the neutron leakage so that the enrichment was increased further.

The sodium void worth and sodium density coefficient are positive because the positive spectral hardening effect outweighs the negative leakage effect. The sodium void worth and sodium density coefficient for the three cores show almost the same values. It was shown that the sodium void worths upon a core plus a fission gas plenum voiding are 6.7$ at a BOEC and 7.3$ at an EOEC for the 600 MWe core and 6.9$ at a BOEC and 7.5$ at an EOEC for the 1 200 MWe core. Those of the 1 800 MWe are 6.9$ at a BOEC and 7.6$ at an EOEC. The sodium density coefficient shows the same tendency. The sodium density coefficients are 0.692 pcm/°C at a BOEC and 0.750 pcm/°C at an EOEC for the 600 MWe core and 0.702 pcm/°C at a BOEC and 0.761 pcm/°C at an EOEC for the 1 200 MWe core. Those of the 1 800 MWe core are 0.697 pcm/°C at a BOEC and 0.761 pcm/°C at an EOEC. The sodium void worth and sodium density coefficient with an increasing power is supposed to increase because of the reduced leakage rate due to an increased core size caused by a higher power. However in this study the active core height was reduced to fix the sodium void worth to be close to 7.5$ so the sodium void worth and the sodium density coefficient have the same value for the three cores.

The radial expansion coefficient represents the reactivity effects of a uniform, radial thermal expansion of the grid plate that is governed by the coolant inlet temperature. The radial expansion coefficient is more negative with a core of a higher power, since the axial leakage increase due to the radial core expansion is larger because of a shorter core height. The radial expansion coefficients are -0.707 pcm/°C at BOEC and -0.743 pcm/°C at EOEC for the 600 MWe core and -0.735 pcm/°C at BOEC and -0.771 pcm/°C at EOEC for the 1 200 MWe core. Those of the 1 800 MWe core are -0.744 pcm/°C at BOEC and -0.780 pcm/°C at EOEC.

The axial expansion coefficient represents the reactivity effects of a uniform, axial thermal expansion of the fuel for the case that fuel is bonded to the cladding. The axial expansion coefficient is less negative with a core of higher power, as the radial leakage increase due to the axial expansion is smaller due to an increased core size. The axial expansion coefficients are -0.160 pcm/°C at BOEC and -0.170 pcm/°C at EOEC for the 600 MWe core and -0.121 pcm/°C at BOEC and -0.127 pcm/°C at EOEC for the 1 200 MWe core. Those of the 1 800 MWe core are -0.109 pcm/°C at BOEC and -0.114 pcm/°C at EOEC.

The radial expansion coefficient increases as the core size increases. However, the axial expansion coefficient decreases as the core size increases at the same rate as the radial expansion coefficient, so the combined effects of the negatively increased radial expansion coefficient and a decreased axial expansion coefficient with a core of a higher power result in minor effects on the reactivity with a higher power. In summary, total reactivity effects with a core of a higher power in the case of considering four effects only, such as the Doppler effect, a uniform axial expansion, a uniform radial expansion and a sodium voiding have little influence on the safety aspects as long as the sodium void worth and the TRU enrichment are calculated to remain at the same value since the increase made by the axial expansion cancels out the decrease made by the radial expansion.

Tables 3, 4 and 5 summarise the heavy metal inventories and mass flow rates of the 600 MWe, 1 200 MWe and 1 800 MWe cores, respectively. Using the TRU feed recovered from three-year cooled LMR spent fuel, the required TRU enrichments were determined such that the same cycle length is achieved. Start-up and recycled core performance characteristics were calculated from REBUS-3 equilibrium cycle analyses. For the start-up core analyses, the LWR-SF TRU was used as TRU feed without recycling the spent fuel. For the recycle core analyses, the recycled TRU from the spent fuel was used as the primary TRU feed, and the LWR-SF TRU was used as the make-up feed.

For the start-up core that only uses the external feed without recycling the spent fuel, the required TRU mass per cycle is 958, 1 886 and 2 796 kg for the 600, 1 200 and 1 800 MWe cores, respectively. The TRU consumption rate is 183 kg/cycle for the 600 MWe core, 367 kg/cycle for the 1 200 MWe core and 543 kg/cycle for the 1 800 MWe core. For the recycled core, the required external TRU mass is reduced to 203 kg/cycle, 388 kg/cycle and 574 kg/cycle for the 600, 1 200 and 1 800 MWe cores, respectively.

Table 3: Heavy metal inventories and mass flow rates of 600 MWe core

	Start-up core					Recycled core				
	Inventory, kg		Mass flow, kg/cycle			Inventory, kg		Mass flow, kg/cycle		
Isotope	**BOEC**	**EOEC**	**Charge**	**Discharge**	**External**	**BOEC**	**EOEC**	**Charge**	**Discharge**	**External**
^{234}U	1.0	1.6	0.0	0.6	0.0	21.2	20.6	4.5	3.9	0.0
^{235}U	19.4	16.1	5.5	2.2	5.5	8.9	8.2	2.1	1.4	0.6
^{236}U	1.6	2.3	0.0	0.6	0.0	8.1	8.1	1.6	1.6	0.0
^{238}U	13 004.2	12 677.2	2 735.3	2 408.3	2 735.3	12 263.4	11 953.1	2 580.3	2 270.0	310.3
^{237}Np	199.3	173.7	51.7	26.1	51.7	93.6	82.7	23.8	12.8	11.0
^{238}Pu	87.3	102.1	9.7	24.4	9.7	154.3	151.3	32.0	29.0	2.0
^{239}Pu	2 182.0	2 071.4	486.7	376.1	486.7	2 069.5	1 966.7	460.1	357.4	103.1
^{240}Pu	974.3	980.4	190.6	196.7	190.6	1 626.2	1 584.3	341.9	299.9	40.4
^{241}Pu	450.1	375.4	128.4	53.7	128.4	311.0	290.5	71.9	51.4	27.2
^{242}Pu	199.6	203.6	37.2	41.2	37.2	361.5	353.7	75.3	67.5	7.9
^{241}Am	123.8	123.0	24.0	23.2	24.0	168.9	157.2	39.0	27.3	5.1
^{242m}Am	4.2	5.5	0.1	1.4	0.1	11.7	11.7	2.2	2.2	0.0
^{243}Am	104.1	97.8	23.8	17.5	23.8	134.0	129.0	29.0	24.0	5.0
^{242}Cm	5.4	6.7	0.0	1.3	0.0	7.4	8.9	0.0	1.6	0.0
^{243}Cm	0.4	0.5	0.1	0.1	0.1	0.8	0.8	0.2	0.2	0.0
^{244}Cm	40.6	46.1	5.3	10.8	5.3	102.6	103.4	19.9	20.7	1.1
^{245}Cm	5.5	7.1	0.5	2.0	0.5	32.1	32.0	6.5	6.4	0.1
^{246}Cm	0.5	0.7	0.1	0.2	0.1	19.2	19.2	3.8	3.8	0.0
Total HM	17 403.4	16 891.1	3 698.9	3 186.6	3 698.9	17 394.4	16 881.5	3 694.1	3 181.2	513.9
TRU	4 377.1	4 194.0	958.1	774.9	958.1	5 092.9	4 891.5	1 105.6	904.2	202.9

Table 4: Heavy metal inventories and mass flow rates of 1 200 MWe core

	Start-up core					Recycled core				
	Inventory, kg		Mass flow, kg/cycle			Inventory, kg		Mass flow, kg/cycle		
Isotope	**BOEC**	**EOEC**	**Charge**	**Discharge**	**External**	**BOEC**	**EOEC**	**Charge**	**Discharge**	**External**
^{234}U	2.0	3.1	0.0	1.1	0.0	39.8	38.7	8.4	7.3	0.0
^{235}U	37.7	31.1	10.7	4.2	10.7	17.2	15.9	4.0	2.8	1.3
^{236}U	3.3	4.6	0.0	1.3	0.0	15.8	15.8	3.1	3.1	0.0
^{238}U	25 449.4	24 797.5	5 358.0	4 706.1	5 358.0	24 836.9	24 200.5	5 229.1	4 592.7	636.4
^{237}Np	389.9	338.5	101.8	50.4	101.8	176.1	155.2	44.9	24.0	20.9
^{238}Pu	173.9	203.5	19.0	48.7	19.0	295.3	289.5	61.3	55.5	3.9
^{239}Pu	4 283.5	4 060.9	958.1	735.5	958.1	4 096.6	3 900.2	907.4	711.0	197.1
^{240}Pu	1 921.0	1 934.0	375.3	388.4	375.3	3 181.3	3 101.0	668.0	587.8	77.2
^{241}Pu	881.9	733.7	252.7	104.5	252.7	604.5	565.5	139.4	100.4	52.0
^{242}Pu	393.7	401.8	73.2	81.4	73.2	695.5	680.5	144.7	129.8	15.1
^{241}Am	241.8	239.3	47.3	44.8	47.3	320.5	298.0	74.1	51.5	9.7
^{242m}Am	8.4	11.1	0.2	2.8	0.2	22.4	22.4	4.3	4.2	0.0
^{243}Am	204.4	191.8	46.8	34.2	46.8	256.8	247.2	55.5	45.9	9.6
^{242}Cm	10.9	13.5	0.0	2.7	0.0	14.3	17.4	0.1	3.2	0.0
^{243}Cm	0.9	1.0	0.1	0.2	0.1	1.6	1.6	0.3	0.3	0.0
^{244}Cm	80.7	91.8	10.4	21.6	10.4	198.9	200.5	38.6	40.2	2.1
^{245}Cm	11.1	14.3	1.0	4.1	1.0	62.4	62.2	12.6	12.4	0.2
^{246}Cm	1.0	1.3	0.1	0.4	0.1	37.5	37.5	7.5	7.5	0.0
Total HM	34 095.4	33 072.8	7 255.0	6 232.3	7 255.0	34 873.3	33 849.6	7 403.4	6 379.7	1 025.6
TRU	8 603.0	8 236.5	1 886.2	1 519.7	1 886.2	9 963.6	9 578.7	2 158.7	1 773.8	387.9

Table 5: Heavy metal inventories and mass flow rates of 1 800 MWe core

	Start-up core					Recycled core				
	Inventory, kg		Mass flow, kg/cycle			Inventory, kg		Mass flow, kg/cycle		
Isotope	BOEC	EOEC	Charge	Discharge	External	BOEC	EOEC	Charge	Discharge	External
^{234}U	3.0	4.7	0.0	1.7	0.0	57.9	56.3	12.3	10.6	0.0
^{235}U	56.1	46.1	16.1	6.1	16.1	25.2	23.2	5.9	4.0	1.9
^{236}U	5.0	6.9	0.0	1.9	0.0	23.4	23.4	4.6	4.6	0.0
^{238}U	38 046.7	37 063.0	8 013.8	7 030.2	8 013.8	37 172.6	36 211.3	7 829.8	6 868.4	961.4
^{237}Np	576.2	499.2	151.0	74.0	151.0	255.7	224.7	65.4	34.5	31.0
^{238}Pu	259.0	303.4	28.2	72.6	28.2	433.8	425.2	90.0	81.4	5.8
^{239}Pu	6 353.1	6 024.8	1 420.1	1 091.7	1 420.1	6 082.1	5 791.6	1 346.7	1 056.2	291.6
^{240}Pu	2 848.6	2 868.6	556.3	576.3	556.3	4 711.8	4 593.1	989.3	870.6	114.2
^{241}Pu	1 303.3	1 082.4	374.6	153.6	374.6	892.0	834.3	206.0	148.3	76.9
^{242}Pu	583.9	596.1	108.5	120.7	108.5	1 024.7	1 002.7	213.3	191.2	22.3
^{241}Am	357.0	352.5	70.2	65.7	70.2	466.9	433.6	108.1	74.8	14.4
^{242m}Am	12.6	16.5	0.3	4.2	0.3	32.8	32.8	6.3	6.2	0.1
^{243}Am	302.6	283.8	69.3	50.5	69.3	377.7	363.5	81.7	67.5	14.2
^{242}Cm	16.3	20.3	0.0	4.0	0.0	21.1	25.6	0.1	4.7	0.0
^{243}Cm	1.3	1.4	0.2	0.3	0.2	2.4	2.4	0.5	0.5	0.0
^{244}Cm	120.2	136.9	15.4	32.2	15.4	294.5	296.8	57.1	59.5	3.2
^{245}Cm	16.6	21.3	1.4	6.2	1.4	92.5	92.3	18.7	18.4	0.3
^{246}Cm	1.5	1.9	0.2	0.6	0.2	55.6	55.6	11.1	11.1	0.0
Total HM	50 862.8	49 329.9	10 825.5	9 292.7	10 825.5	52 022.7	50 488.3	11 046.9	9 512.4	1 537.3
TRU	12 752.1	12 209.2	2 795.7	2 252.7	2 795.7	14 743.6	14 174.1	3 194.3	2 624.8	574.0

Conclusion

To achieve a core power level as high as possible without penalising core safety, a large monolithic sodium-cooled fast reactor for a TRU burning with a power ranging from 600 MWe to 1 800 MWe was designed. With the design data accumulation accompanied by the safety analysis, the results of this study would be able to identify the most limiting factor for scaling-up burner core concepts.

For a TRU burner core design, the design approach of a variable cladding thickness and a variable core height which adjusted the active height to make the sodium void worth close to 7.5$ was used. Core design used the design constraints related to the current technology database with a TRU enrichment limit (30.0 w/o) and a fast neutron irradiation limit (4.0×10^{23} n/cm^2). A single enrichment concept was adopted to flatten the power distribution and fuel pin designs of different cladding thicknesses were used for different core regions. For the core design concepts, three power levels of 600, 1 200 and 1 800 MWe were selected. For a consistent comparison, the number of assemblies was adjusted to attain a similar linear power of around 180 W/cm. The fuel pin diameter and the number of fuel pins were fixed to 7 mm and 271 respectively. But, the pitch to diameter of the fuel rods was allowed to vary. In order to demonstrate a high TRU consumption rate, a core design was evaluated to attain a TRU enrichment of 30 wt.% as much as possible. The core design was confirmed in that the maximum inner cladding temperatures are below 650°C and the maximum pressure drops are below 0.15 MPa.

The calculation results show that large monolithic sodium-cooled fast reactors for a TRU burning whose power range is from 600 to 1 800 MWe can be designed satisfactorily. The burn-up reactivity swing can be retained at around 3 500 pcm and the consumption rate is increased at almost the same rate as the increased power so it seems to have no preference at any power level with the same TRU enrichment. But a core with a higher power has an disadvantage from the reactivity aspect due to its increased sodium density coefficient with an increased core size. However, in this study, it was shown that this increased sodium density coefficient can be improved if the pancake core option is adapted so the active core height was adjusted to make the sodium void worth close to the same value. It can thus be said that the total reactivity effects with a core of a higher power in the case of considering those effects only, such as the Doppler effect, a uniform radial expansion, a uniform axial expansion and sodium voiding have little influence on the safety aspects as long as the sodium void worth and the TRU enrichment are calculated to remain at the same value since the increase made by the axial expansion cancels out the decrease made by the radial expansion. The only disadvantage with regard to scaling-up the burner core concepts is an increased effective core radius to conserve the same linear power due to a core height reduction for a reduction of the sodium void reactivity.

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Study of minor actinide transmutation in sodium fast reactor depleted uranium radial blanket

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Abstract

This paper presents the last results obtained of minor actinide transmutation in a sodium fast reactor depleted uranium radial blanket. This concept is based on a heterogeneous multiple recycling model. The use of the oxide matrix allows to reprocess such S/A in the spent fuel standard flow.

The major advantages are that this concept has a small impact on the safety coefficient, has no impact on core management and enables longer irradiation time for minor actinides giving rise to high transmutation rates. For the study, we use a preliminary design of a 3 600 MWth sodium fast reactor in progress at CEA. We investigate the transmutation performances of $(U+Np+Am+Cm)O_2$ *fuel in radial blankets assemblies. We focus on two upper and lower assumptions in order to investigate the feasibility domain for this concept: one with a minor actinide (MA) content of 10%, and the second with an enrichment of MA close to 40%. The study shows that it is possible to reach good transmutation rates with these two concepts. But because of the high content of minor actinides, we observe a high specific heat for the assembly loaded with MA, a high decay heat level and also a high neutron source level, in particular for the 40% MA content. We show that investigations are necessary to propose a R&D programme to take into account the assembly manufacturing, the assembly manipulation, the transportation of these assemblies, the fuel reprocessing and to consider the fuel behaviour under irradiation.*

Introduction

In the framework of next-generation fast reactor design, the management of minor actinides (MA) is one of the key issues. The transmutation of MA can be achieved with various modes of transmutation and waste management. Two means of transmutation are possible:

- the homogeneous method, wherein the minor actinides to be transmuted are directly mixed with the "standard" fuel of the reactor;
- the heterogeneous method, for which the actinides to be transmuted are separated from the fuel itself, in limited number of S/A (targets) devoted to actinide transmutation.

Associated with two methods for actinide management:

- Multiple recycling: in this case all or part of the minor actinides and plutonium at the end of each reactor cycle is sent back in the following cycle. In this way, only reprocessing losses go to the ultimate waste.
- Once-through: in this case the minor actinides are transmuted in targets where very high burn-up is reached.

The principle of the transmutation consists in recycling waste in the nuclear reactors while seeking for MA to favour their fission ("final destruction") compared to the neutron capture (which does nothing but move the problem…). Fast reactors offer the best performances to transmute the minor actinides in a homogeneous or heterogeneous way at industrial scale. The safety criteria are acceptable for whole solutions if the MA content is not over 2.5% of the total heavy nuclides [1]. An alternative way consists of a decoupled fuel/actinide management by loading minor actinides with oxide matrix in radial blankets of a sodium fast reactor. The main advantages are the following:

- This model has only small impact on sodium void coefficient (few %)
- It has almost no impact on the core management.
- It enables longer irradiation time for minor actinides giving rise to high transmutation rates.
- The introduction of actinides degrades plutonium quality as compared to traditional uranium blankets, which might facilitate the verification of proliferation resistance criteria.

Despite all these advantages, this concept meets sharp challenges that have to be carefully considered for the assembly design:

- high specific heat for assembly manufacturing;
- high decay heat level for in-core and out-of-core assembly manipulation;
- high neutron source level (fuel reprocessing);
- fuel behaviour under irradiation (swelling and helium production).

The results of the process of designing a heavily loaded minor actinide blankets assembly placed at the periphery of a sodium fast reactor are presented and the performance of transmutation of minor actinides involved. A previous study [2], based on the design of the European Fast Reactor (EFR), had already shown the interest of decoupled fuel processing plutonium and minor actinides on the main characteristics of the core (safety coefficients, fuel management, etc.). Here, we will implement this study on a first image of a Gen-IV sodium fast reactor design by the CEA. We focus on two upper and lower assumptions in order to investigate the feasibility domain for this concept: one with a minor actinide (MA) content of 10%, and the second with an MA enrichment close to 40%.

Design process

The design of such an assembly has to meet all these constraints and needs to go beyond solely neutronic considerations. To do so, we settled a multi-physic process that can comply with most of the corresponding criteria. It is based on an iterative process that couples neutronics, fuel thermal behaviour and thermal-hydraulic calculations. Obtaining performance transmutation requires a realistic design of the sub-assembly in realistic operating conditions of this new type of core. Based on the irradiation characteristics in the radial blanket, it seeks a design assembly respecting all of the technological constraints already identified in the EFR case. The design is based on an iterative process summarised in Figure 1.

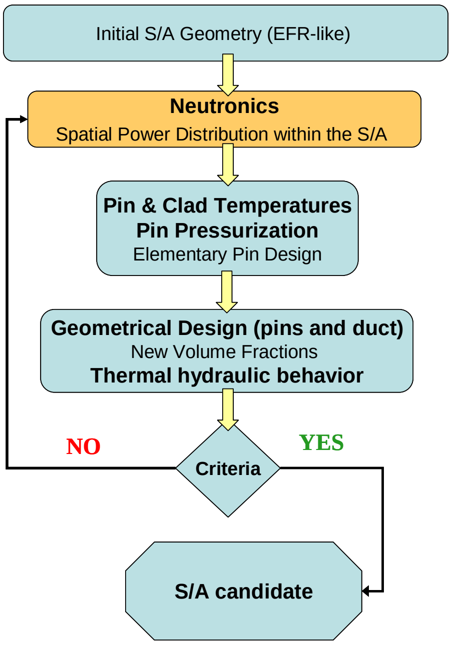
Neutronic calculation scheme

Calculations were performed using the ECCO/ERANOS [3] code package together with an adjusted ERALIB1 library which provides accurate predictions for sodium-cooled fast reactors. Applications to innovative designs would have to be verified but is probably sufficient in this preliminary design phase. The transport option has been used for all calculations with a 33-group library which has been generated from a 1 968-group master library. For comparison, fine-group (1 968 groups) ECCO cell calculations were also performed, and provide an accurate description of the reaction thresholds and resonances. The fine-group calculation has been performed for the two-dimensional description of a sub-assembly. Because the broad 33-group library has been generated from this master fine group library, it was found that differences on the core parameters are small. For the geometry model of the core, a cylindrical (RZ) model is used. Validity of the RZ model was assessed against the results of heterogeneous (hexagonal-Z) calculation using the TGV/VARIANT three-dimensional nodal transport code. Considering the core layout, the radial blanket location is on the fourteenth row (84 S/A outer the core). The description of the corresponding blanket volume was split into nine regions of equal volume in order to extract spatial dependence of the pertinent variables used in the physical analysis (damage, helium production and burn-up). Heavy nuclei depletion calculations are performed with an extended chain up to ^{252}Cf isotope.

Iterative design process

Starting from an existing radial blanket S/A design, neutronics calculations were performed to provide relevant data to the other disciplines. These are the inputs needed to check the viability of the current candidate. The pressurisation analysis and the thermal behavior (fuel and cladding) aim at optimising the fuel fraction inside the bundle and at determining which of the two materials candidates (traditional AIM1 steel and promising oxide dispersion strengthened steel) would be more suitable for the cladding.

Figure 1: Iterative process description



For the two scenarios (10 and 40% MA in the radial blankets), the methodology (described in Figure 1) has been applied using the spatial pin distribution across the assembly: it has been shown that the pin set close to the core/blanket interface undergoes the most severe conditions in each case. To optimise the fuel fraction inside the bundle, the pin bundle geometry is considered and slight corrections to the pin outer diameter are proposed. In case of significant modifications either on the fuel pellet outer diameter or on the induced cladding stress values, the entire procedure has to be performed from the beginning. At the end of these iterative calculations, acceptable pin and pin bundle geometry is obtained from a thermo-mechanical point of view. Finally, a complete design is proposed. If the obtained volume fractions are quite similar to the initial neutronic input, the S/A can be considered to be appropriate for such a SFR core. Otherwise, the process is performed until convergence is reached.

Core and MA blanket description and management

Core description

The main objective was to obtain an innovative SFR based on oxide fuel that can reduce the sodium void effect in a significant way together with a positive (but near zero) internal breeding gain. Reducing the sodium volume fraction and increasing the fuel volume fraction is highly attractive for the three following key variables: sodium void effect, Doppler effect and breeding gain. The first core characteristics are summarised in Table 1.

Table 1: Assembly and core characteristics and SFR

Assembly and core characteristics	SFR	
Total thermal power (MWth)	3 600	
Pellet and hole diameter (mm)	9.5	2
Spacer wire diameter (mm)	1	
Number of pins per assembly	271	
Grid pitch (mm)	206	
Fuel volume fraction (%)	47.4	
Sodium volume fraction (%)	27.1	
Structure volume fraction (%)	18.5	
Fissile length (cm)	100	
Sodium plenum length (cm)	30	
Total core height (m)	1.30	
Number of fuel assemblies	424	
Fuel zone volume (m^3)	15.58	
Total fertile volume (m^3)	0.00	
Fuel zone diameter (m)	4.6	
Max. linear power (W/cm)	450	
Average core power density (W/cm^3)	231	

Figure 2: Fuel S/A

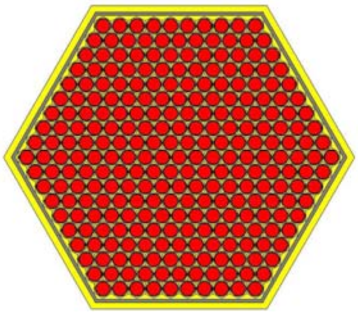
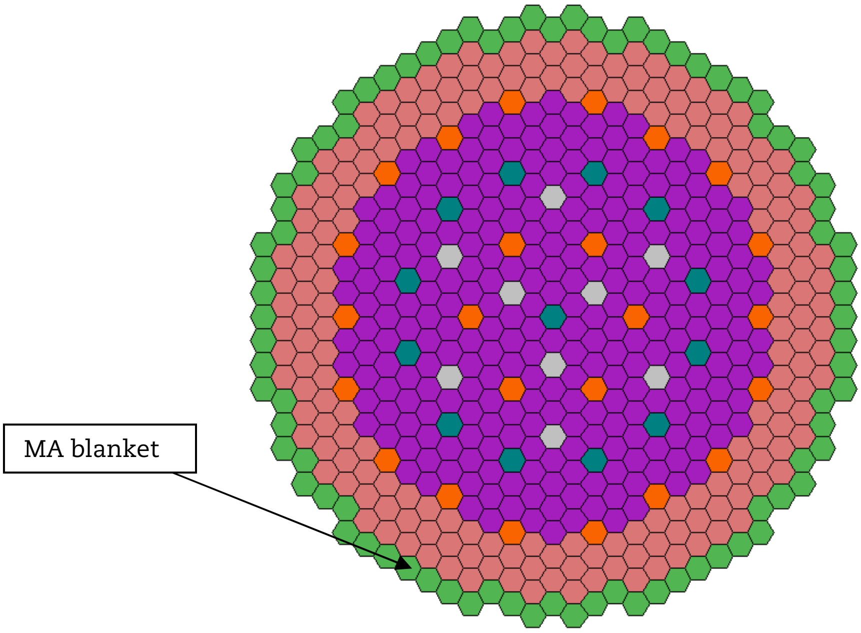


Figure 3: Core layout



The core is managed based on a five multi-batch scheme simulated by a single full batch scheme. This means that all assemblies are discharged at the same time at the end of life but that the reactivity over the period of time between 2/5 and 3/5 of the residence time is maintained positive. The cycle length is then 1/5 of the total fuel residence time which is considered limited by a maximum damage of 200 dpa on the cladding.

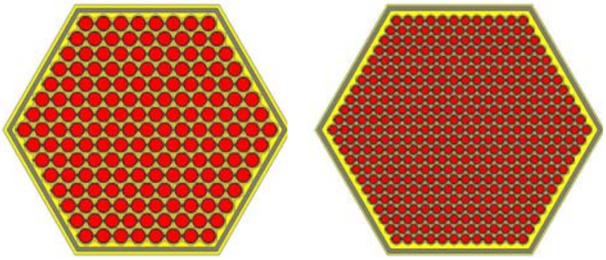
Blanket description

The design of the radial blanket S/A is driven by the iterative process. The fuel is a mixture of U and minor actinide (MA) oxide. Here, we consider two different cases:

- an ambitious 40% MA content that is intended to be close to what is expected for accelerator-driven system fuel;
- a 10% MA content easily achievable by near-term technology.

The first case enables to maximise the MA loaded mass in the blankets and may lead to a small part of the power plant fleet using blankets to achieve a MA equilibrium (production of the whole fleet = destruction in the radial blankets). It is the most attractive case and will be considered as our main objective. The second one is a more realistic view of the problem in which all SFR cores have radial blankets. The process starts with the original EFR radial blanket design (volume fractions), but with a reduced fuel length of 100 cm to deal with the core fuel assembly design.

Figure 4: MA blanket S/A 10% and 40%



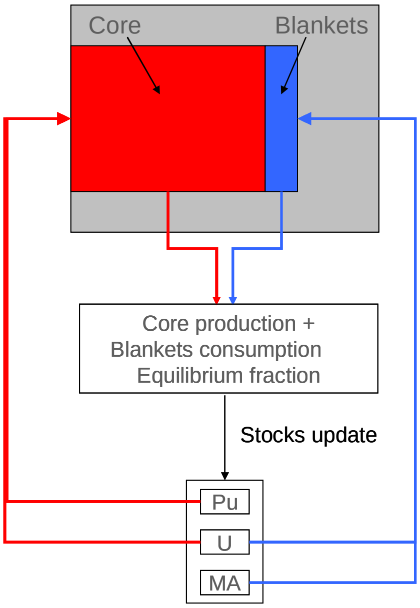
Minor actinide management

The multi-recycling process has been performed in order to obtain an equilibrium core. One of the major goals of this particular heterogeneous model is to concentrate minor actinides in radial blankets and to ensure minor actinide stability for the entire power plant fleet. To accomplish this goal, some basic hypothesises are necessary:

- The fleet is a mixture of the same SFR core with and without radial blankets.
- The part of SFR with blankets is determined by both MA consumption in the blankets and MA production in the core.
- The core is loaded with 100% $(U,Pu)O_2$ fuel. The Pu enrichment is adjusted with respect to fixed cycle length.
- The blankets are always reloaded with the same initial $(U+MA)O_2$ mass.
- The burned heavy nuclei mass of the blankets is filled with depleted uranium and MA coming from the core production.
- The SFR cycle length is 5 × 410 equivalent full power days, whether it has blankets or not.

Figure 5 shows a synoptic description of the mutli-recycling.

Figure 5: Heterogeneous multi-recycling scheme



Results

Core performances

The intrinsic performances of the core are detailed in Table 2.

Transmutation performances

Table 3 shows the assembly mass balance for 40% and 10% cases for equilibrium core. For both cases, the transmutation rate is close to 41%. The fraction of cores with blankets in the power plant fleet to ensure MA equilibrium is 23% for the 40% MA case and 88% for the 10% MA case. Assuming a 40% thermal efficiency, the MA consumptions in the blanket reach -12 kg/TWeh and -3.5 kg/TWeh respectively (the MA production of the fuel core is +3.2 kg/TWeh).

Table 2: Core performances for SFR (without blanket)

Core performances	SFR
Total thermal power (MWth)	3 600
Fuel life time (EFPD)	2050
Maximum burn-up (GWd/t)	184
Mean burn-up (GWd/t)	106
Mean Pu content (%)	14.7
Initial Pu mass (t)	10.5
Mean breeding gain	+0.04
Fuel voiding end of life ($)	5.4
Fuel voiding + plenum end of life($)	4.4

Table 3: Transmutation performances for the equilibrium core

Concept	40% MA		10% MA	
Mass inventory (BOL and EOL)	Charged (kg)	Discharged (kg)	Charged (kg)	Discharged (kg)
U	79.0	71.2	130.9	118.3
Pu	0.0	17.4	0.0	12.6
Np	9.0	5.1	2.6	1.5
Am	35.8	18.5	9.8	5.0
Cm	8.0	7.6	2.2	2.1
H. N.	131.8	119.9	145.5	139.5
Transmutation rate	40.9%		41.1%	
Fraction with blanket	23%		88%	
MA consumption	-12 kg/TWeh		-3.5 kg/TWeh	

In both cases, we note curium equilibrium. In this typical fast neutron spectrum, the $\sigma_{capture}/\sigma_{fission}$ ratio of ^{244}Cm is close to 1. Under irradiation, Am isotopes transmute into ^{242}Cm and ^{244}Cm after series of neutron captures and beta decay. The ^{242}Cm isotope quickly decays into ^{238}Pu + α, while ^{244}Cm disappears by fission or is transmuted into ^{245}Cm which is fissile. The obtained curium mass shows a large ^{244}Cm contribution (~80%) for both charged and discharged cases. Concerning the plutonium production in the blankets, we can see in Table 4 that, whatever the case, the isotopic content shows a high level of ^{238}Pu and ^{240}Pu. The ^{238}Pu comes from ^{242}Cm which is continuously produced by ^{241}Am isotope under irradiation, with a very short half-life (164 days). ^{240}Pu is also produced from alpha decay of ^{244}Cm but with a larger half-life (18 years). For the 10% MA case, the high level of ^{239}Pu comes from the higher amount of ^{238}U mass available for neutron capture than in the 40% MA case.

Table 4: Isotopic content of the reprocessed plutonium produced in the radial blankets

Isotope	40% MA	10 % MA
^{238}Pu	46	23
^{239}Pu	39	65
^{240}Pu	15	12

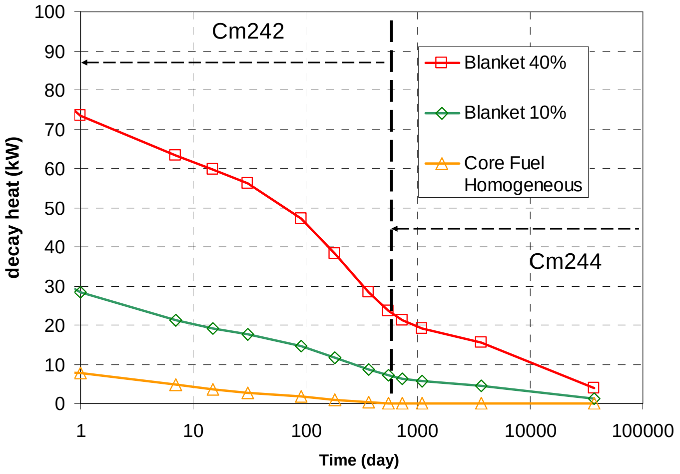
These results show good insights for non-proliferation issues for the 40% MA case and even for the 10% one. The 84 radial blankets assemblies around the standard fuel core produced 672 kg of ^{238}Pu each 4 100 EFPD while 605 kg are discharged from the fuel core in the same time. When combined with the 23% fraction of the cores using blankets, the ^{238}Pu part of the plutonium in the stock goes from 2.1% to 3.2%. The large part of americium and especially curium in the blanket leads to some problematic challenges for the fuel cycle front end: manufacturing, storage and transportation of a fresh fuel assembly. The challenging issue is the thermal power and neutron source of the fresh fuel assembly. Table 5 shows the values for the 40% and 10% cases as compared to the homogeneous MA multi-recycling for the same SFR core.

Table 5: Thermal power and neutron source for fresh S/A

	SFR homogeneous (UPu + 0.7% MA)	Radial blanket 40% MA	Radial blanket 10% MA
Thermal power (kW)	0.7	21.6	5.4
Neutron/s	$1.7\ 10^{9}$	$8.0\ 10^{10}$	$1.9\ 10^{10}$

As shown previously, the heat level is driven by the curium content in the assembly. The same problem arises for the residual heat power (decay heat). As the curium mass remains stable in case of MA equilibrium, the decay heat is at least equal to the fresh fuel thermal power which is already high for back-end problematic. In addition, the ^{241}Am transmutation produces ^{242}Cm isotopes that remain stable under irradiation (competition between creation and disappearance by alpha decay). This contribution has a large impact on the decay heat behavior. Figure 6 shows the time dependence of decay heat for the 40% MA and 10% MA as compared to the standard fuel with homogeneous multi-recycling. In the first ten days of cooling, the decay heat is driven by the fission products. From 10 days to two years the decay heat comes mainly from ^{242}Cm alpha decay. Then the ^{244}Cm is the only contributor (more than 98%) due to its large half life (18 years). The three systems behave basically the same way. The only change is the curium amount.

Figure 6: Time dependence of the decay after irradiation



The 40% MA and 10% MA decay heat cases reach their initial fresh fuel thermal heat level after three years of cooling. However, the 10% MA case exhibits a lower decay heat. Considering an optimistic value of 40 kW as a threshold below which assemblies can be removed from the active zone of the core, the standard fuel with homogeneous multi-recycling and the 10% MA case would be handled within three months after the power shutdown. In the 40% MA case, specific equipments have to be studied in order to deal with a high heat level such as 50 kW to handle these assemblies. Investigations are under way at CEA to proposed R&D toward such systems.

Moderated blanket

In order to increase transmutation performances a promising method is to locally moderate the fast spectrum. The locally moderated spectrum allows increasing locally the cross-section while maintaining the intrinsic high-level flux of the fast reactor and finally increases the reaction rate [4]. A preliminary evaluation of the transmutation performances (see Table 6) with a zirconium hydride moderator allows reaching a high transmutation rate (~70% vs. 40% for the non-moderated blanket) during the same irradiation time. The level of the neutron source increases by a factor 100, and the impact will be evaluated.

Table 6: Transmutation performances for moderated vs. fast blanket

Type of spectrum blanket	Fast	Moderated
Moderator fraction ($ZrH_{1.78}$) (%)	0.00	47
Heavy nuclides – S/A (kg)	145	35
MA mass – S/A (kg)	14.6	14.2
MA transmutation rate	41%	74%
Np transmutation rate	43%	85%
Am transmutation rate	49%	90%
Cm transmutation rate	3%	16%
Am fission rate	20%	37%
MA consumption (kg/TWe)h	-3.5	-6.2
S/A thermal power (BOL)	5.4	4.3
S/A heat decay (EOL + 1 year)	8.6	6.3
S/A neutron source (EOL + 2 years)	$1.9\ 10^{10}$	$4.7\ 10^{12}$

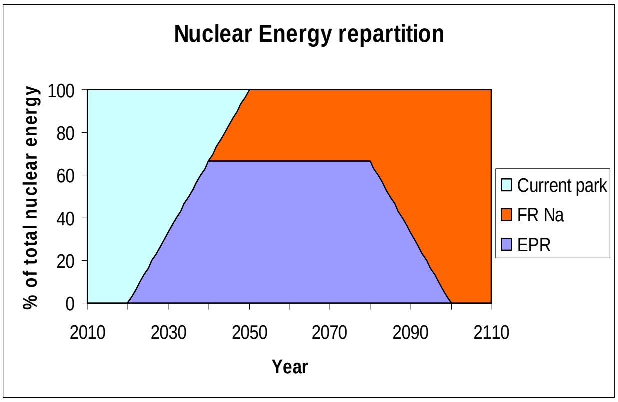
Scenario

The CEA is studying scenarios of principle for the French case through a dynamic vision of the nuclear development, based on the one hand on the solutions offered by the existing fleet (reprocessing, fabrication and NPP) and on the other hand on the solutions offered by the future fourth-generation reactors. This scenario is declined then according to Figure 6. The scenario study considers the French nuclear park with a constant nuclear energy demand at 430 TWhe/year. The current nuclear park is replaced between 2020 and 2050 by a mixed nuclear park: 67% of Generation III EPR reactors and 33% of Generation IV SFR. From 2080 to 2100, the EPR are replaced by SFR. The plutonium is recycled in the fissile part of the SFR core. The separation of the minor actinides at the reprocessing step starts in 2038. The minor actinides are recycled in the radial blankets of the SFR from 2040 (10% MA content). Those calculations are performed by the COSI code [5]. The results indicate that the minor actinide inventory can be stabilised with the heterogeneous mode of transmutation using minor actinides in the radial blankets of the SFR. The initial MA fraction in the radial blankets necessary to completely stabilise the minor actinide inventory in the fleet is linked to many assumptions or results, including:

- The stabilisation of the plutonium inventory: an increase of the Pu inventory induces an increase of the americium inventory due to the decay of ^{241}Pu in the spent fuel storage.
- The cooling time of the spent fuel before reprocessing, for the same reasons.
- The recycling of the reprocessed uranium in the fast reactors, which induces an extra production of neptunium in the fast reactors.

For these reasons, an initial minor actinide fraction from 10-20% in the radial blankets is sufficient to stabilise the minor actinide inventory. The optimisation of the transmutation scenarios is ongoing.

Figure 6: Scenarios with fast reactor deployments



Conclusion

The minor actinides multi-recycling on a depleted uranium oxide matrix in radial blankets of SFR showed good results in terms of transmutation performances. This heterogeneous model allows a massive minor actinide loading while having almost no consequence on the core safety parameters and core fuel management. Two MA enrichment targets have been studied: an ambitious 40% case and a more realistic 10% case. The design of such an assembly must deal with criteria implying multi-physics analysis. The 10% MA content seems to achieve a good balance between transmutation performance and back/front end impact (neutron source, decay heat, etc.) as compared to the 40% content. Another promising means of increasing the transmutation performance is the local moderation blanket. The investigations, such as dedicated experimental material and fuel irradiation programmes, are ongoing at CEA to set a global vision of an optimised system that can answer all these questions.

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Closing session

Chair: K. Minato

Workshop summary

A closing session was held at the end of the meeting. K. Minato (Japan), the Chair of the Information Exchange Meeting, gave a summary of the event. A total of 153 participants from 14 countries and 3 international organisations participated. A total of 117 presentations 117 were given (45 oral presentations and 72 posters). The number of participants in alphabetical order by country is: Belgium (1); Canada (2); Czech Republic (2); France (13); Germany (8); India (1); Italy (5); Japan (75); Korea (12); Portugal (2); Russian Federation (4); Spain (5); Sweden (8); United States (9); EC (3); IAEA (1); NEA (2). The number of presentations by session is:

- Opening session – NEA, IAEA and EURATOM activities: three oral presentations.
- Special session – Fuel cycle strategies and transition scenarios: eight oral presentations and two posters.
- Session I – Impact of P&T on waste management and geological disposal: three oral presentations and three posters.
- Session II – Transmutation fuels and targets: eight oral presentations and seven posters.
- Session III – Partitioning, waste forms and management: ten oral presentations and 34 posters.
- Session IV – Materials, including spallation targets and coolants: four oral presentations and two posters.
- Session V – Transmutation physics experiments, nuclear data: three oral presentations and six posters.
- Session VI – Transmutation systems: six oral presentations and 18 posters.

An additional special session, “Fuel cycle strategies and transition scenarios”, was held for the first time and met with success. There was a general consensus that the topic was well chosen and relevant. France, Japan and the USA have stable national strategies concerning the advanced fuel cycle, and the equilibrium transition scenario model has shown good improvement. The NEA initiated transition scenario studies on both regional and global cases have been progressing well. Scenarios concerning P&T technology maturity should be carried out in terms of national and global transition scenarios. Transition scenarios should take into account the impact on geological disposal and a closer relationship needs to be developed with the experts on geological disposal.

Following a suggestion from K. Minato, technical sessions were summarised by co-chairs based on the following four questions:

- What are the notable achievements or what progress has been made in each research area?
- What is to be done for further development?
- What are the technical obstacles to further development?
- What is recommended for the development of P&T technology?

Notable achievements and progress made in P&T

- P&T’s impact on geological disposal studies, *e.g.* RED-IMPACT;
- Irradiation behaviour of fuels containing minor actinides and their interaction between cladding materials.

- The basic properties of minor actinide materials.
- Alternative partitioning processes, *e.g.* chromatographic extraction and crystallisation methods.
- Better understanding of the chemistry of minor actinide separation in both aqueous and pyro-processes.
- Great progress made in the area of uncertainty analysis of nuclear data: sensitivity analysis technique and covariance data.
- Progress made in related facilities, experiments and international projects: MEGAPIE, TEFT, J-PARC, GUINEVERE, KUCA, YALINA, etc.
- Compared to previous editions, remarkable progress achieved as pertains to critical and subcritical transmutation systems design (Session VI), a sign of moving toward the industrialisation of P&T.

Needs for further development

- Political and financial support for the successful implementation of industrial-scale P&T.
- An effort to develop and select low activation materials that reduces intermediate-level waste and secondary waste streams in P&T processes, *e.g.* waste minimisation.
- Curium management.
- Demonstration of minor actinide fuel fabrication and irradiation testing of minor actinide fuels in target burners.
- Alternative technology in solvent extraction method should be developed [current: amides, DGA, BT(B)P method].
- Study on the integration of the partitioning processes in “reference” to fuel cycle scenarios.
- Study on the accountability or traceability of nuclear data errors.
- Enhance differential measurement technology for minor actinides and Pu isotopes.
- More experiments simulating transmutation systems, both critical and subcritical.
- Demonstration of subcritical system with spallation source coupling.
- Study on practical applications of P&T technologies and transmutation systems.
- Economic and cost benefit studies.

Technical obstacles

- Limited demonstration facilities: the “hot” laboratories.
- Lack of reliable fast neutron irradiation system, *e.g.* Phénix, Joyo.
- Insufficient human and financial resources.
- Difficulties in industrial-scale implementation: minor actinide fuel fabrication, partitioning processes and transmutation systems.
- Remote handling of minor actinide fuel fabrication due to high decay heat and radiation of fabrication: slowdown of industrialisation.
- Measurement, sample preparation and handling of minor actinide data are difficult.
- Cost of building and managing P&T facilities are too high.
- Transportation of radioactive materials and spent fuels for P&T-related issues.

Recommendations for future P&T technology

- The geological disposal community should accept that P&T is a viable option in radioactive waste management.
- Take into account natural decay time of nuclear material before setting up a P&T roadmap.
- Simulation and modelling studies of fuel fabrication and reprocessing technologies.
- Sharing fuel cycle and testing facilities.
- Simplifying and optimising the existing partitioning processes.
- Demonstration of P&T feasibility on an industrial scale.
- Early optimisation and selection of systems and fuel cycle options.
- Validation of licensing and operation procedures.
- Critical and subcritical experiments using minor actinide fuels will be the next step in realising transmutation systems.
- Continue international collaborations and cross-cutting programmes in the field of P&T.
- Maintaining a close relationship with industry and utilities that will design the system and operate the fuel cycle facilities; this will ease industrial implementation of P&T.
- Summarise twenty years of the NEA's effort on P&T (*e.g.* P&T handbook). This will help identify needs and a way of drawing-up a long-term plan for P&T and the advanced fuel cycle as well as persuading the public and decision makers.

K. McCarthy (United States) announced the intention to host the 11th Information Exchange Meeting in the second half, in the US. There also was a proposal from the Czech Republic to host the 12th Information Exchange Meeting in 2012.

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