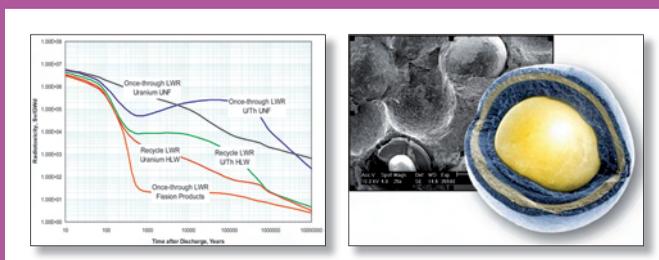


# Actinide and Fission Product Partitioning and Transmutation

Eleventh Information  
Exchange Meeting  
San Francisco, California, USA  
1-4 November 2010





## **Actinide and Fission Product Partitioning and Transmutation**

### ***Eleventh Information Exchange Meeting***

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NUCLEAR ENERGY AGENCY  
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

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- 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
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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 in advanced fuel cycles considering both waste management and economic issues.

In order to provide experts a forum to present and discuss state-of-the-art developments in the P&T field, the OECD/NEA has been organising biennial information exchange meetings on actinide and fission product partitioning and transmutation since 1990. The previous meetings were held in Mito (Japan) in 1990, at Argonne (United States) 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 (United States) in 2004, in Nîmes (France) in 2006 and in Mito (Japan) in 2008. They have often been co-sponsored by the European Commission (EC) and the International Atomic Energy Agency (IAEA).

The 11<sup>th</sup> Information Exchange Meeting was held in San Francisco, California, United States on 1-4 November 2010, comprising a plenary session on national P&T programmes and six technical sessions covering various fields of P&T. The meeting was hosted by the Idaho National Laboratory (INL), United States.

The information exchange meetings on P&T form an integral part of NEA activities on advanced nuclear fuel cycles. An overview of NEA activities on P&T and relevant publications are available at [www.oecd-nea.org/pt/](http://www.oecd-nea.org/pt/).

These proceedings include all the papers presented at the 11<sup>th</sup> 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 Idaho National Laboratory (INL) for hosting the 11<sup>th</sup> Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation. It also extends its gratitude to the European Commission (EC) and the International Atomic Energy Agency (IAEA) for their co-operation.

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Poster session contributions are available at [www.oecd-nea.org/pt/iempt11/](http://www.oecd-nea.org/pt/iempt11/)

## Executive summary

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

Eleven information exchange meetings have been organised thus far (Mito, Japan in 1990; ANL, United States in 1992; Cadarache, France in 1994; Mito, Japan in 1996; Mol, Belgium in 1998; Madrid, Spain in 2000; Jeju, Korea in 2002; Las Vegas, United States in 2004; Nîmes, France in 2006; Mito, Japan in 2008; and San Francisco, United States in 2010). This 11<sup>th</sup> meeting was hosted by the Idaho National Laboratory (INL) 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: fuel cycle strategies and transition scenarios; radioactive waste forms; the impact of P&T on geological disposal; radioactive waste management strategies (including secondary wastes); transmutation fuels and targets; pyro and aqueous separation processes; materials, spallation targets and coolants; transmutation physics, experiments and nuclear data; transmutation systems (design, performance and safety); handling and transportation of transmutation fuels; and economics of P&T. These topics encompass subjects such as multi-scale modelling, experiments and instrumentation, novel theories and benchmarks. A plenary session was organised to present national programmes related to P&T.

A total of 102 presentations (42 oral and 60 posters) were discussed by 131 participants from 17 countries and 3 international organisations. A closing session with a panel discussion was also organised at the end of the meeting.

### Opening and Plenary Session: National Programmes on P&T

#### ***Chairs: Th. Dujardin (NEA), K. McCarthy (INL, United States)***

The opening session began with welcome addresses from Kathryn A. McCarthy, Deputy Associate Laboratory Director of Idaho National Laboratory and Thierry Dujardin, Deputy Director of the OECD Nuclear Energy Agency. These were followed by seven presentations, three of which from international organisations and four from different countries.

- Th. Dujardin (NEA) gave an overview of ongoing NEA activities regarding actinide and fission product partitioning and transmutation. Numerous and varied activities related to P&T are currently being conducted by the NEA, covering both scientific and strategic aspects. The main activities in the areas are transition scenarios, fuel and materials, and fuel cycle chemistry. Recently, increased interest has been directed towards the study of the thorium cycle, the nuclear fuel “deep burning” concept, and the economics of nuclear power. The need to consider industry views was also addressed.
- The IAEA Co-ordinated Research Project (CRP) on analytical and experimental benchmark analyses of accelerator-driven systems was presented by A. Stanculescu (IAEA). The IAEA Technical Working Group on Fast Reactors (TWG-FR) aims to improve the physics of subcritical systems and to provide data and codes to enhance related R&D programmes. The scope of the CRP is to perform computational and experimental benchmarking on

ADS and non-spallation neutron source driven subcritical systems. Introductions to the following work domains were given: YALINA Booster; Kyoto University Critical Assembly (KUCA); Pre-TRADE; FEAT (First Amplifier Tests); TARC (Transmutation by Adiabatic Resonance Crossing); ADS kinetics analytical benchmarks.

- On behalf of G. Van Goethem (EC), P. D'Hondt (Belgium) gave a presentation on the EURATOM research and training programme for P&T. The EU has been supporting various P&T related activities since 2007 under the auspices of the Framework Programme 7 (FP7). The European Sustainable Nuclear Energy Technology Platform (SNE-TP) was set up to help make decisions related to demonstration facilities to be built at a 2015-2020 time horizon. Two collaborative projects, ACSEPT, which studies partitioning technologies and actinide science and EUROTRANS, which studies transmutation of high-level nuclear waste in ADS, were summarised.
- The United States' nuclear energy research and development and the move towards a long-term, science-based, goal-oriented programme was presented by C. Savage (DOE, United States). The DOE Office of Nuclear Energy and R&D roadmap were introduced. The objectives of the United States' nuclear energy R&D are: life extension, building new nuclear facilities, development of a sustainable fuel cycle and minimisation of proliferation risk. The direction of the United States' future nuclear fuel cycle was also summarised.
- D. Warin (CEA, France) summarised future nuclear systems for France, specifically fuel cycle options and guidelines for research. The recent situation of French nuclear energy status was reported. Currently 850 HM of spent nuclear fuel is being recycled per year, and a total of 25 kHM has now been reprocessed in France. The French long-term strategy is based on implementing fast reactor systems, and a prototype reactor, ASTRID, will be constructed to operate from 2025. Related reprocessing technologies were also discussed.
- Minor-actinide-bearing fuel study activities in Japan for a homogeneous TRU recycling fast reactor system was presented by T. Mizuno (JAEA, Japan). The current status of the FaCT project, the fast reactor cycle development programme in Japan, and the Monju reactor was reported. For FaCT fuel development, the Monju reactor was restarted using Am-bearing fuels. Further development will be promoted by the Joyo reactor. International collaboration with the United States and France will continue.
- On behalf of K-C. Song (KAERI, Korea), J-G. Kim (KAERI, Korea) presented perspectives of pyroprocesses for long-term spent fuel management in Korea. The main goal of the Korean nuclear strategy on spent fuel management is to develop proliferation-resistant nuclear energy. Hence, under the framework of the national nuclear R&D programme, sodium-cooled fast reactor with pyroprocessed nuclear fuel was selected. Korea is also preparing a high-level waste disposal system and decontamination and decommissioning of existing facilities.

## **Session I: Fuel Cycle Strategies and Transition Scenarios**

***Chairs: T. Mizuno (JAEA, Japan), B. Dixon (INL, United States)***

This session comprised six oral presentations:

- An international comparison for transition scenario codes involving COSI, DESAE, EVOLCODE, FAMILY and VISION was presented by L. Boucher (CEA, France). This programme was performed as a benchmark activity under the OECD/NEA Expert Group on Fuel Cycle Transition Scenario Studies. Since the codes have different output and capabilities, the goal of the benchmark is to assess the common capabilities of the codes. The main outputs and conclusions were presented.

K. Ono (JAEA, Japan) reported on the result of global scenarios for fast reactor deployment. The scenario is based on a homogeneous scenario with a MOX-fuelled fast reactor, which

is being developed by Japan. Two representative scenarios were studied with different assumptions concerning fuels, deployment time, breeding ratio, etc. Both scenarios were compared using the cumulative natural uranium demand, reprocessing capacity, nuclear power generation, etc.

- N. Camarcat and C. Garzenne (EDF, France) presented on industrial research for transmutation scenarios in EDF. The EDF has been performing transition scenarios using SFR to manage various minor actinides. The benefits and drawbacks of P&T were analysed, especially as linked to the notion of geological repository. Due to its high impact, the study mainly focused on Am transmutation and implementation in advanced nuclear fuel cycles. The conclusion showed that the total Am transmutation may reduce the total repository area by as much as 30% (in 2150).
- B. Petrovic (Georgia Tech, United States) discussed a requirements-driven comprehensive approach to fuel cycle back-end optimisation. Westinghouse and Georgia Tech are currently developing a requirements-driven approach to devising solutions for the fuel cycle back-end. The advantages and drawbacks of a thorium nuclear fuel cycle and related technologies were summarised.
- The topic of a world transition towards a sustainable nuclear fuel cycle was presented by V. Romanello (KIT, Germany). Both homogeneous and heterogeneous recycling methods were evaluated based on a transition from thermal to fast reactor systems. The conclusion shows that the deployment pace of fast reactors in the different world regions is determined by various factors such as future energy demand, resource availability, fast reactor conversion ratio, reprocessing and fabrication capacities, etc.
- An overview on homogeneous and heterogeneous transmutation in a new French SFR and the supposed reactor and fuel cycle impacts was given by F. Varaine (CEA, France). Based on a 2006 French law, CEA is developing the SFR prototype ASTRID, which also aims at minor actinide transmutation. For fuel development both homogeneous and heterogeneous methods are being considered. The presentation focused on the recent R&D status of both recycling methods.
- H. Oigawa (JAEA, Japan) presented on the current status and future plans for research and development on partitioning and transmutation technology in Japan. R&D on P&T technology has shown significant progress in Japan. A second “Check and Review” (C&R) was conducted by JAEC in 2008-2009. The evaluation of current technology levels of P&T showed that most of the related technologies are still at the basic research level due to the lack of experimental data for minor actinides.

Ten additional papers were presented during the poster session.

## **Session II: Waste Forms and Geological Disposal**

***Chairs: W.M. Nutt (INL, United States), I-S. Hwang (SNU, Korea)***

This session comprised four oral presentations:

- Fuel cycle potential waste for disposition was discussed by R. Jones (SRNL, United States). The presentation covered estimation and analysis of characteristics of various US used fuels (UNF) originated from commercial and military facilities. The used fuels consist of UNF from LWR, second wastes from LWR recycling processes, used MOX and secondary waste from recovering TRU.
- P. Swift (SNL, United States) summarised recent R&D on repository safety assessments done by the United States, Sweden, France, Switzerland and the European Commission. The study considers three topics: radionuclides contributing to total dose, thermal loading and waste form lifetime. The conclusion indicated that  $^{129}\text{I}$  is the major contributor due to its long half-life.

- R. Wigeland (INL, United States) discussed the relationship between geologic disposal characteristics and advanced nuclear fuel cycles with partitioning and transmutation. The study showed that P&T can be effective in improving nuclear waste management for used fuel and high-level waste, and the uncertainty dominates predictions of repository performance.
- J-H. Yoon (UC Berkeley, United States) introduced the environmental impacts of the Korean advanced nuclear fuel cycle KIEP-21 and disposal concepts. The study focused on the radionuclide transport model and environmental performance analysis based on the Korean reference spent fuel disposal system (KRS) and advanced KRS (A-KRS) which considers pyroprocessing of UNF.

In addition, three papers were presented during the poster session.

### **Session III: Transmutation Fuels and Targets**

***Chairs: K. Minato (JAEA, Japan), K. Pasamehmetoglu (INL, United States)***

This session comprised six oral presentations:

- F. Delage (CEA, France) discussed minor actinide transmutation in a prototype accelerator-driven system, and the results from fuel developments within the European EUROTRANS programme. The presentation focused on the AFTRA project, which develops fuel for European ADS. The recent outcomes of cermet and cermet fuel element design and performance assessments, safety analyses and in/out-of-pile experiments were reported.
- H. Ohta (CRIEPI, Japan) reported on the development of minor actinide transmutation by CRIEPI. CRIEPI studies the development of metallic fuel for fast reactors. Details concerning reprocessing methods, irradiation test experiments, performance analysis and quantitative analysis were presented.
- The concept and development programmes regarding the minor-actinide-bearing blanket at CEA was presented by J-M. Bonnerot (CEA, France). Post-irradiation examination is expected to take place in 2012, as are semi-integral irradiation experiments.
- G. Bell (ORNL, United States) summarised the subject of fuels for the United States DOE Deep Burn programme. Tristructural isotropic (TRISO) particle fuel is being developed for high-temperature gas-cooled reactors. “Deep burn” refers to the large fractional TRU burn-up that can be achieved in a TRISO-fuelled gas-cooled reactor. The current status of R&D and future plans were described.
- Progress concerning the CEA ALFA project (ATALANTE Laboratory for Actinide-bearing Fuels) was reported by V. Royet (CEA, France). The ALFA project aims to develop fuel for the ASTRID fast reactor which is planned in France. The roadmap for the R&D programme, fuel fabrication technologies, shielding issues and design studies were summarised.
- The behaviour of zirconium on passivated U-Zr-based fuel alloys was presented by R. Mariani (University of Las Vegas, United States). The presentation covered the range of zirconium behaviours, potential problems for Zr-alloy fuels, ternary U-Pu-Zr alloys cast and examinations, sample preparation methods, XRD Rietveld refinement results, preliminary SEM results and future plans.

In addition, 11 papers were presented during the poster session.

## Session IV: Pyro and Aqueous Processes

**Chairs:** *D. Warin (CEA, France), J. Uhlíř (NRI, Czech Republic)*

This session comprised eight oral presentations, of which two were invited papers:

- S. Bourg (CEA, France) discussed the ACSEPT project, concerning progresses in advanced partitioning and challenges for the future. The ACSEPT project aims at the development of P&T technologies through multi-national collaboration between 12 European countries, Japan and Australia. The current status of the project and future plans were summarised.
- Y. Morita (JAEA, Japan) presented on the development of separation technology at JAEA with regard to transuranium elements and fission products through the use of new extractants and adsorbents. Various separation steps for TRU extraction, Cs and Sr separation as well as future plans were reported.
- D. Warin (CEA, France) summarised recent progress with advanced actinide recycling processes in France. The presentation covered innovative minor actinide partitioning hydrometallurgical processes and the results of related technologies such as Ganex, Diamex, Sanex, ExAm and Tedga. Milestones for future development were also presented.
- One-cycle Sanex process development studies and laboratory-scale demonstrations were the subjects addressed by A. Wilden (FZJ, Germany). The FZJ is conducting research on the Sanex process within the framework of the ACSEPT project. Details of the process, experimental results, flow sheets and future plans were presented.
- The development of a novel Ganex process was summarised by E. Aneheim (University of Chalmers, Sweden). The Ganex process is one of the separation processes being considered for future nuclear systems. Chalmers University is developing a Ganex process through the combination of two extractants: BTBP and TBP. The results of the development process were addressed.
- G.D. DelCul (ORNL, United States) presented on the subject of the advanced head-end for treatment of used LWR fuel. The main concept of the novel dry head-end process is separated cladding and removal of upfront fission products. Details of the process and the experimental results were summarised.
- J. Uhlíř (NRI, Czech Republic) gave an overview on current R&D progress on pyrochemical partitioning technology in the Czech Republic. The P&T programme in the Czech Republic mainly aims towards implementing MSR/MSTR with fuel fabricated by the fluoride volatility method. The flow sheet and development detail were discussed.
- A pyropartitioning experiment to recover TRU from high-level liquid waste was presented on by K. Uozumi (CRIEPI, Japan). The main advantages of the pyrochemical process are to minimise proliferation risk and reduce secondary wastes. Development details such as denitration, chlorination and reductive-extraction were addressed.

In addition, 17 papers were presented during the poster session.

## Session V: Transmutation Physics and Materials

**Chairs:** *E.M. González-Romero (CIEMAT, Spain), C. Fazio (KIT, Germany)*

This session comprised five oral presentations:

- The first presentation of the session was a review of Pb alloy coolant chemistry and structural material performance by C. Fazio (KIT, Germany). Coolant chemistry control, handling and material compatibility issues were summarised. The result will be published in the revised NEA handbook on heavy liquid metal.

- Critical analysis of the effective delayed-neutron fraction calculation routes using deterministic and Monte Carlo codes was presented by M. Carta (ENEA, Italy). The presentation covered theoretical analysis, comparison on standard and More Carlo  $\beta_{\text{eff}}$  formulations, ERANOS calculation and results and future plans.
- E.M. González-Romero (CIEMAT, Spain) summarised a study on experimental validation of reactivity monitoring techniques for power ADS systems to incinerate radioactive wastes. The experiment was performed for the Yalina booster subcritical assembly. Related technologies such as PNS, beam trip and recent development progress were presented.
- F-J. Hambsch (JRC-IRMM, Belgium) presented on reaction cross-sections, fission yields and prompt neutron emission from actinide targets. The presentation addressed topics such as neutron-induced fission reaction ( $^{234}\text{U}$ ), neutron emission in fission of  $^{252}\text{Cf}$  (SF), prompt fission neutron spectrum of  $^{235}\text{U}(n_{\text{th}},f)$  and  $^{241}\text{Am}$  transmission and capture cross-sections.
- Investigation of nuclear data accuracy for the accelerator-driven system with minor actinide fuel was summarised by K. Nishihara (JAEA, Japan). The study was performed within the framework of the IAEA CRP benchmark activity on depletion analysis for ADS. The uncertainty analysis using covariance matrix was presented.

In addition, four papers were presented during the poster session.

## Session VI: Transmutation System: Design, Performance and Safety

**Chairs: P. D'Hondt (SCK•CEN, Belgium), A. Stanculescu (IAEA)**

This session comprised six oral presentations:

- C-H. Pyeon (Kyoto University, Japan) gave an overview of the progress made as concerns accelerator-driven systems in the Kyoto University Critical Assembly (KUCA). Two experimental test results and future plans were presented,  $^{235}\text{U}$ -loaded ADS experiments with 14 MeV neutrons and  $^{235}\text{U}$ - and  $^{232}\text{Th}$ -loaded ADS experiments with 100 MeV protons.
- K. Minato (JAEA, Japan) presented the notion of technology readiness levels (TRL) for partitioning and transmutation of minor actinides in Japan. The definition of the TRL has evolved so as to evaluate R&D status for Japanese P&T technology. The study examined current TRL for the Japanese minor actinide transmutation systems, partitioning processes and minor-actinide-bearing fuels.
- Advanced fuel cycle scenario study in the European context using different burner reactor concepts was summarised by V. Romanello (KIT, Germany). The presentation was focused on the European advanced nuclear fuel cycle and its scenarios using ADS. The fusion system was also considered for the scenario study.
- H. Ait Abderrahim (SCK•CEN, Belgium) gave an overview of the MYRRHA project, which is the multi-purpose hybrid research reactor for high-tech applications. The development status of the MYRRHA system was summarised. The strategy for implementing a P&T fuel cycle and future plans were presented as well.
- F. Venneri (LOGOS Tech., United States) presented a contribution on the Deep Burn project, providing an overview of the progress made. The United States' Deep Burn programme, concept and various fuel cycle scenarios were summarised. Recently, development on TRISO fuel as well as reactor design study showed good progress.
- T.K. Kim (ANL, United States) presented a systematic evaluation of uranium utilisation in nuclear systems. The presentation contained information on uranium utilisation versus fuel cycle options, advanced once-through fast systems and assessment of uranium utilisation. Possible sustainable fuel cycles were summarised in conclusion.

In addition, nine posters were presented during the poster session.

## Closing Session

### **Chair: K. McCarthy (INL, United States)**

A closing session was held wherein session co-chairs were invited to present the highlights from their technical sessions and to form a panel for an open discussion with the attendees.

The main conclusions and recommendations of the meeting are as follows:

- Strong international collaborations and progress in developing cross-cutting programmes in the field of P&T should be continued.
- The role of P&T technology becomes more important as a means of waste management and effective use of energy resources, which helps establish a sustainable nuclear fuel cycle and leads to the reduction of greenhouse gas emissions.
- Study of the economics of P&T and cost-benefit analysis of a P&T embedded nuclear fuel cycle scenario, which consists of a reduction of decay heat and radiotoxicity, are necessary.
- P&T scenario uncertainty must be mitigated, particularly as regards long-term geological disposal scenarios.
- Transport issues should be studied.
- P&T-related nuclear data and the level of its uncertainty should be studied.
- Facilities for irradiation testing, fast spectrum testing, innovative fuel demonstration testing, pyro/aqueous processing development, etc., remain insufficient.
- A modelling study on actinide chemistry is needed.
- Industrialisation of P&T technologies, pyro/aqueous processes, innovative fuels, fast nuclear systems, etc., are needed.
- Knowledge management and maintenance of accumulated information is necessary.

J. Uhlír (NRI, Czech Republic) announced that NRI would be hosting the 12<sup>th</sup> Information Exchange Meeting (12-IEMPT) in the second half of 2012, in the Czech Republic.

The meeting was closed by K. McCarthy, the Chair of the Scientific Advisory Committee.



# Opening



## INL welcome address

**K. McCarthy**

Idaho National Laboratory  
Idaho Falls, ID, United States

On behalf of Idaho National Laboratory, it is my honour to welcome you to the 11<sup>th</sup> International Exchange Meeting on Partitioning and Transmutation. As you know, this series of meetings has been organised by OECD Nuclear Energy Agency since 1990 to provide a forum to discuss current issues and R&D needs for future development of partitioning and transmutation. This is the third time that the United States has hosted the IEMPT; the second IEMPT was hosted by Argonne National Laboratory, and the eighth IEMPT was hosted by the University of Nevada, Las Vegas. The INL is pleased to have the opportunity to host the 11<sup>th</sup> IEMPT.

In the first IEMPT, the focus was mainly on the chemistry and physics of partitioning and transmutation (P&T). Over the last ten years, many international collaborative activities have begun, such as the Generation IV International Forum, the Global Nuclear Energy Partnership, INPRO and others. As a consequence, the scope of the IEMPT has evolved to cover scientific and strategic issues of P&T as well as industrialisation and deployment of advanced nuclear fuel cycles.

In the United States, there is much attention directed towards nuclear fuel cycle activities. The Blue Ribbon Commission on America's Nuclear Future has been commissioned by the US Secretary of Energy, at the request of the President, to conduct a comprehensive review of policies for managing the back-end of the nuclear fuel cycle and recommend a new strategy. The US Department of Energy Fuel Cycle R&D Program is evaluating fuel cycle options against various criteria and conducting a broad range of R&D to support that evaluation. The Idaho National Laboratory, the US Department of Energy's lead laboratory for nuclear energy research, is involved in all aspects of fuel cycle development including advanced fuels R&D development, separations R&D, proliferation risk reduction, advanced reactor design and fuel cycle systems analysis. The United States will continue to work with its international partners to explore fuel cycle options, and this series of meetings is an important venue for presenting and discussing fuel cycle R&D.

This year we have accepted more than 120 abstracts from various OECD/NEA member states, with topics covering the scientific and strategic/policy developments in the field of P&T, including:

- fuel cycle strategies and transition scenarios;
- radioactive waste forms;
- impact of P&T on geological disposal;
- radioactive waste management strategies (including secondary wastes);
- transmutation fuels and targets;
- pyro and aqueous separation processes;

- materials, spallation targets and coolants;
- transmutation physics, experiments and nuclear data;
- transmutation systems: design, performance and safety;
- handling and transportation of transmutation fuels;
- economics of P&T.

It is my hope that this 11-IEMPT will help to foster new international and national collaborations, further P&T knowledge, and move us closer to deployment of an advanced nuclear fuel cycle.

As chair of this meeting, I'd like to thank you again for joining us in San Francisco. I hope that you can take some time to enjoy this beautiful city and its surroundings while you are broadening your technical network and knowledge.

Thank you very much.

## OECD/NEA welcome address

**Th. Dujardin**

OECD Nuclear Energy Agency  
Issy-les-Moulineaux, France

On behalf of the OECD Nuclear Energy Agency, it is a great honour for me to welcome you all to the 11<sup>th</sup> Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation.

First of all, I would like to express my sincere appreciation to our hosts, the Idaho National Laboratory and the chair of the meeting, Dr. Kathryn McCarthy, the Deputy Associate Laboratory Director for Nuclear Science and Technology of INL, as well as to all INL staff and the members of the scientific advisory committees, who have worked hard in preparing and supporting this meeting.

I would also like to thank the European Commission and the International Atomic Energy Agency for their co-operation in the organisation of this meeting.

This year is the 20-year anniversary of this meeting. In 1988, based on the Japanese government initiative long-term R&D 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 partitioning and transmutation. This series of information exchange meetings is one of the outcomes of this international project on P&T. The objective of this meeting is to provide a forum for experts to present and discuss the results of their research with colleagues.

When we first met in Mito, Japan in November 1990 there were only about 20 presentations covering chemistry and physics of P&T. After the year 2000, the size of the meeting greatly increased and its scope has been extended to cover scientific and technological aspects of advanced nuclear fuel cycles as well as related strategies and policy issues. This year, about 150 papers will be presented either in oral or poster sessions. The 11<sup>th</sup> meeting is the largest meeting ever in terms of number of papers and participants. I am very pleased to see that the interest in P&T is still high after 20 years of activity.

The 11<sup>th</sup> meeting will cover various topics of P&T including fuel cycle strategies and transition scenarios, radioactive waste forms, impact of P&T on geological disposal, radioactive waste management strategies, transmutation fuels and targets, pyro and aqueous separation processes, materials, spallation targets and coolants, transmutation physics, experiments and nuclear data and transmutation systems design, performance and safety. Following the decision of the related standing NEA committee in 2007, every second meeting there will be a special session dedicated to a specific field of research, replacing the reports on national programmes. At last meeting in Mito in 2008, the special session on fuel cycle strategies and transition scenarios was organised and this year a report on national programmes on P&T will be given later this morning.

As you well know, the main energy policy these days focuses on secure supply of resources, enhanced economics and environmental protection. In this framework, nuclear energy could be a viable option for future sustainable energy policy as well as to achieve the “green growth”

strategies which the OECD is pursuing. Last year was the 50<sup>th</sup> anniversary of the NEA and there was a ministerial-level meeting to discuss the future of nuclear energy for peaceful use and challenges for new nuclear build. In connection with this anniversary, we published a *Nuclear Energy Outlook* to provide comprehensive analysis on current and future nuclear industry and recommendations on the challenges to be faced. At the conclusion of these activities, we addressed many of the political and scientific issues related to the so-called nuclear renaissance. For example, governmental or inter-governmental policies on nuclear issues should be stabilised, economics and safety should be enhanced, high-level waste repositories should be implemented, social acceptance should be improved, qualified human resources should be secured, Generation IV reactor technologies should be demonstrated successfully, etc.

Partitioning and transmutation embedded advanced fuel cycles may offer benefits of reducing radiotoxicity and the burden of the radioactive wastes, which is one of key challenges for future sustainable nuclear systems. According to a recent NEA-IEA joint publication, *Technology Roadmap – Nuclear Energy*, in order to achieve successful deployment of advanced fuel cycles, the governments should continue to support R&D of advanced nuclear technology, the international community should continue to strengthen co-operation on the development of advanced reactor and fuel cycle technologies, and the nuclear industry and utilities should participate, in co-operation with nuclear research institutes, in the development of next-generation nuclear systems to ensure that the designs selected for demonstration are those most suitable for eventual commercialisation.

For the last two decades, we watched the great progress in the P&T related technologies. The Generation IV International Forum is in good shape and many countries are expanding their effort on developing various advanced nuclear systems. I hope this 11<sup>th</sup> Information Exchange Meeting will be meaningful for all the participants presenting on the state of art of P&T technology, and will enhance further international collaborations.

Before closing my welcome address, I am happy to announce that the NEA has received a proposal from the Nuclear Research Institute in the Czech Republic to host the next information exchange meeting on P&T in the autumn of 2012 in Prague. The details will be announced in due course.

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 San Francisco.

Thank you for your attention.

## **Plenary Session**

### **National Programmes on P&T**

***Chairs: Th. Dujardin, K. McCarthy***



## OECD/NEA activities related to partitioning and transmutation

**Th. Dujardin, Y-J. Choi**  
OECD Nuclear Energy Agency  
Issy-les-Moulineaux, France

### Abstract

*For more than 20 years, the OECD Nuclear Energy Agency (NEA) has been conducting international studies related to the partitioning and transmutation (P&T) of nuclear waste, including physics, chemistry and material issues, 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 Science Committee (NSC) and the Nuclear Development Committee (NDC), are part of this programme with the objective of enhancing 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. Recently, the NEA reviewed the potential benefits and impacts of advanced fuel cycles with partitioning and transmutation, as well as national programmes in chemical partitioning. The NEA has also compared criteria for a choice between homogeneous and heterogeneous recycle modes and studied the impact of P&T on potential radiological storage and repositories. This paper provides an overview of results from recent NEA activities in the field of P&T and provides an insight into ongoing projects and planned future activities.*

## Introduction

OECD/NEA P&T-related activities have been carried out under the auspices of the Nuclear Science Committee (NSC) and the Nuclear Development Committee (NDC), in order to cover both scientific and strategic issues. The NSC's work is mainly done by expert groups of the Working Party on Scientific Issues of the Fuel Cycle (WPFC), including nuclear fuel cycle transition scenarios, impact of P&T on radioactive waste management, innovative fuels and materials and fuel cycle chemistry and minor actinide management. The work of the NDC mainly consists of strategic and assessment reports covering the broader field of P&T and its impact on the economics of the nuclear fuel cycle, such as trends in the nuclear fuel cycle. The OECD/NEA also directs the peer review on a P&T-related national programme: the MYRRHA project (Belgium).

Organising the Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation has been one of most eminent achievements of P&T-related OECD/NEA activities. The meeting was initiated in 1989 following a proposal from the Japanese government which aimed for international collaboration and information sharing on issues related to P&T. Since the first meeting held in Mito, Japan in 1990, the scope of the meeting has been expanded from basic P&T technology to advanced nuclear system design. Today, implementation and industrialisation of P&T are under discussion. The event is held every two years in different NEA member countries. The 12<sup>th</sup> meeting, hosted by NRI, will be held in September 2012 in Prague, Czech Republic. The NEA also organises two P&T-related workshops, one on "Technology and Components of the Accelerator-driven Systems (TCADS)", held in Germany in 2010, and one on "Structural Materials for Innovative Nuclear Systems (SMINS)", held in Korea, also in 2010.

## Nuclear fuel cycle issues

In many of countries long-term sustainable nuclear systems are under development, and there are various technical and strategic issues when adapting such systems. The transition from current thermal nuclear systems to future fast nuclear systems could be quantified by studying scenarios based on energy demand, resources required and technical maturity, as well as political parameters. Since 2005 the NEA has been working in this field to discuss related issues and identify R&D needs. Three publications were delivered in 2009:

- a status report on fuel cycle transition scenarios that provides a framework for assessing national needs regarding fuel cycle transition and covers the timing of key technologies;
- a regional study on potential European fuel cycle transition to examine implementation of the innovative fuel cycles associated with partitioning and transmutation in Europe;
- strategic and policy issues raised by the transition from thermal to fast nuclear systems which covers political and economic issues of the transition scenarios.

## ***Benchmark of transition scenario codes***

In order to quantify the transition scenarios, computer simulation codes have been developed to model the dynamic transition from an initial state to a final state for nuclear energy. The results are mainly the mass flow and the composition of heavy elements depending on time: natural uranium needs, enrichment needs, fresh fuel fabrication needs, fuel irradiation, inventory of spent fuel and nuclear materials, and reprocessing needs as well as nuclear waste production, economical aspects and non-proliferation criteria. Since 2007 the NEA has been conducting a benchmark on these computer codes to compare capabilities, modelling methods and results. Two phases of calculation were performed: depletion and transition modes. Three fuel cycle scenarios were studied: once-through cycle; limited Pu recycling in LWR cycle; and Pu and minor actinide recycling in fast reactor cycle. Codes from Belgium, Canada, France, Germany, Italy, Japan, Spain and the United States were used. The report will be published in 2011.

### ***Global transition scenarios***

The NEA also performed a scenario study based on the global perspectives for future fuel cycles with fast reactors, using both a homogeneous and a heterogeneous approach of the different world regions. The heterogeneous approach did consider a subdivision of the world into four main macro-regions based on the countries' economic development status. An original global electricity production envelope was used in simulations, and a specific regional energy share was defined. In the regional approach two different fuel cycles were analysed: the once-through LWR cycle was used as the reference, and then a transition to a fast reactor closed cycle for a better management of resources and minimisation of wastes. The main objective in both cases (homogeneous or heterogeneous world approaches) was to deploy fast units as quickly as possible and to replace the thermal reactor fleet by the fast reactor one in order to minimise the uranium resource consumption and to cope with increasing global world energy demand. The analysis on the infrastructure requirements as a function of time and a sensitivity study was also performed. The report will be published in 2011.

### ***Trends in the nuclear fuel cycle***

The renewed interest in the environmental and energy security advantages of nuclear energy has led to a spur of new thinking in the development of technologies such as Generation IV and P&T. The advent of Generation III, III+ and IV reactors will require changes in fuel fabrication, reprocessing, waste conditioning, etc. With much happening in connection with nuclear fuel cycle development, an update of the 2002 NEA publication *Trends in the Nuclear Fuel Cycle: Economic, Environmental and Social Aspects*, is being undertaken. The main objective of the work is to review the previous publication by investigating the trends in the latest progress and the future trajectories of the nuclear fuel cycle. The scope of the study will cover a comprehensive review of existing publications and the assessment of advancements in relevant technologies, ongoing global, international initiatives, progress and programmes of individual countries, with in-depth analyses of specific case studies. Whilst trends in the aforementioned features – from the past to today, to what will be required tomorrow – will be underlined from a technological perspective, particular emphasis will be placed on policy and strategy issues and their reciprocal influence on the course of technological progress, as well as sustainability aspects associated with the nuclear fuel cycle, such as long-term supply of fuel resources, radioactive waste management and potential links to nuclear proliferation. The report will be published at the end of 2011.

### ***Potential benefits and impact of advanced fuel cycles with P&T***

Several impact studies have been performed over the last few years that have underlined the role of radiotoxicity or heat load and their potential reduction. Hence, the NEA conducted comparative analysis studies which were performed in several international laboratories on the impact of advanced fuel cycles including partitioning and transmutation (P&T) on geological repository performance. This analysis assesses the impact on geological repository performance and characteristics, and indicates possible goals for future studies. Further, it drafts recommendations on the appropriate criteria to evaluate P&T impact, on the level of losses at fuel processing, etc. P&T allows a reduction of the inventory of the emplaced materials that can have a significant impact on the repository, even if not overwhelmingly high. Moreover the inventory reduction can have the effect of making the uncertainty about repository performance less important both in normal evolution and in particular in case of strongly disruptive scenarios. It is also emphasised that, while P&T will never replace the need for a deep geologic repository, it has the potential to significantly improve the public perception of the ability to effectively manage radioactive wastes by largely reducing the transuranic waste masses to be stored and, consequently, to improve public acceptance of a geologic repository. Both issues are important for the future sustainability of nuclear power. The report will be published in 2011.

## **Nuclear fuels and material**

In this field the NEA has been continuously updating information on fuels and materials for implementing advanced nuclear fuel cycles. Activities cover evaluation of innovative fuels and materials technologies, including fabrication processes and performances. Study of heavy liquid metal (HLM) technology is also performed through publication of a series on heavy liquid metal, e.g. lead-bismuth eutectic, handbook.

### ***Innovative fuels***

The NEA is preparing a status report on innovative fuels to study technical issues associated with the development of innovative fuels, mostly minor actinide containing fuels and clad materials, targeted for use in advanced fuel cycles. The report covers various fuel types such as metal, oxide, nitride, dispersion fuels (cercer, cermet) and special mechanic fuel forms (sphere-pac, vibro-pac, particle fuels). The above fuels will be assessed in terms of fabrication technology, characterisation and irradiation performance, safety characteristics, design and safety criteria, system dependency with cladding materials and technical readiness level.

### ***Innovative structural materials***

A status report is being prepared on the innovative structural materials under extreme conditions such as high temperature, high dose rate, corrosive chemical environment and long service lifetime. Technical issues concerning gas-cooled fast reactor (GFR), very high-temperature reactor (VHTR), sodium-cooled fast reactor (SFR), lead-cooled fast reactor (LFR), molten salt-cooled reactor (MSR) and supercritical water-cooled reactor (SCWR) as well as the accelerator-driven systems (ADS) are being summarised.

## **Nuclear fuel cycle chemistry**

As regards nuclear fuel cycle chemistry the NEA focuses on the subjects of the technical assessments of various separations processes, developing a scientific basis for nuclear waste recycle, and nuclear data for radioactive waste management. In 2010, the NEA published 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, the Russian Federation and the European Commission were summarised. A total of 12 spent nuclear fuel recycling flow sheets of the various reprocessing processes were collected in order to evaluate critical performance parameters for each process and scenario. Management and separation technologies related to Cm, a high toxicity waste, have also been studied.

### ***Progress in separation chemistry and future R&D***

The goal of this activity is to update information on reprocessing technologies. The study summarises progress in separation technology and current achievements, including hydrometallurgy, pyrometallurgy (metal and oxide electro-refining), fluoride volatility and head-end processes. The separation requirements from the viewpoint of fuel cycle scenarios as well as the perspective of future R&D will also be assessed.

### ***Integral experiments for minor actinide (MA) management***

The management of minor actinides (MA) such as neptunium, americium and curium in the spent fuel, renders important the use of mixed-oxides (MOX) or adapting P&T concepts in the nuclear fuel cycles. In order to minimise the uncertainty of physical parameters when loading MA, accurate MA nuclear data are indispensable. Compared to the nuclear data of major actinides, e.g. uranium and plutonium, data on MA are few since the integral experiments on MA are more difficult due to insufficient facilities, difficulty of preparing samples and repetition of

tests, the need to improve measurement techniques, etc. Since 2009 the NEA has therefore been conducting a critical review on integral experiments for validating MA nuclear data, as well as to recommend additional integral experiments needed for validating MA nuclear data and to investigate the possibility of establishing an international framework to facilitate integral experiments for MA management. The first draft of the report will be ready by the end of 2011.

### ***International peer review of the MYRRHA project***

In 2009, at the request of Belgian government, the NEA performed an international peer review on the MYRRHA project, an accelerator-driven lead-bismuth eutectic cooled subcritical reactor. The review was done by a team of seven high-level experts from seven different countries. As a conclusion of the review, “risk reduction and allocation before proceeding to construction” is recommended. The Belgian government will fund 40% if the interest of potential investors translates into tangible commitments.

## **Conferences and workshops**

### ***Workshop on Technology and Components 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. Taking into account the suggestion from the fifth workshop in Belgium in 2007 (HPPA5), the NEA decided to change the title and scope of the workshop to cover the development status of accelerator-driven systems (ADS): Workshop on Technology and Components of ADS (TCADS). The first TCADS workshop was held on 15-17 March 2010 in Karlsruhe, Germany and was hosted by KIT. A total of 70 presentations (oral and poster) were given by about 100 of participants. The workshop focuses on current R&D status of ADS, neutron sources, application of ADS and system design issues. The conclusions of the workshop are as follows:

- Various programmes around world are progressing, including EUROTRANS (EC), MYRRHA (Belgium), J-PARC (Japan), SNS (United States) and MEGAPIE (Switzerland).
- Key technologies such as beam stability, windowless target, spallation sources and materials have been improved.
- Design studies of subcritical systems are increasing, considering fuel cycle issues, fuels and materials, and safety analysis.
- Innovative use of ADS is also being examined, e.g. ADS with thorium fuels, Am/Cm-dedicated burning ADS.

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

The second SMINS workshop was held on 31 August-3 September 2010 in Daejeon, Korea, hosted by KAERI. About 80 papers were presented and more than 100 experts participated. The scope of the workshop covers fundamental and applied studies on metals, ceramic and novel materials. Multi-scale modelling issues were discussed as well. In the summary session, “Critical Experiments, Modelling Needs and Round Robin Opportunities”, participants agreed upon the following conclusions:

- Issues for innovative structure materials were identified.
- Technical readiness level (TRL) could be a proper tool to evaluate the current R&D level.
- An international database on structural materials for innovative nuclear system is needed.
- Further collaboration with materials scientists in non-nuclear fields is needed.

***Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation***

Since 1990, the NEA has been organising a forum for the discussion of scientific and strategic developments in the field of P&T. For the 11<sup>th</sup> meeting, more than 140 abstracts were submitted from 13 countries and 3 international organisations. The abstracts covered the following topics: fuel cycle strategies and transition scenarios; waste forms and geological disposal; transmutation fuels and targets; pyro and aqueous separation processes; transmutation physics and materials; transmutation systems: design, performance and safety. As an opening session, national programmes on P&T are reported. The 12<sup>th</sup> Information Exchange Meeting will be held during the second half of September 2011 in the Czech Republic, hosted by NRI.

**Concluding remarks**

The OECD Nuclear Energy Agency (NEA) carries out numerous and various activities in the field of partitioning and transmutation (P&T), covering both scientific and strategic aspects. Current main activities focus particularly on nuclear fuel cycle transition scenarios, fuels and materials development, and fuel cycle chemistry. Interest in the thorium fuel cycle and the nuclear fuel “deep burn” concept has increased. The P&T community is very vital and though P&T a large number of nuclear fuel cycle options can be taken into consideration. For future implementation, industrialisation should be examined.

## The IAEA Co-ordinated Research Project (CRP) on “Analytical and Experimental Benchmark Analyses of Accelerator-driven Systems”\*

**Alexander Stanculescu**

*(on behalf of all CRP participants)*

Division of Nuclear Power  
Nuclear Power Development Section  
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### Abstract

Since December 2005, the International Atomic Energy Agency (IAEA) has been conducting the Co-ordinated Research Project (CRP) on “Analytical and Experimental Benchmark Analyses of Accelerator-driven Systems” within the framework of the Technical Working Group on Fast Reactors (TWG-FR).

The overall objective of the CRP is to increase the capability of interested member states in developing and applying advanced reactor technologies in the area of long-lived radioactive waste utilisation and transmutation.

The specific objective of the CRP is to improve the present understanding of the coupling of an external neutron source (e.g. spallation source) with a multiplicative subcritical core. The participants are performing computational and experimental benchmark analyses using integrated calculation schemes and simulation methods. The CRP aims at integrating some of the planned experimental demonstration projects of the coupling between a subcritical core and an external neutron source [e.g. YALINA Booster in Belarus, and Kyoto University’s Critical Assembly (KUCA)]. The objective of these experimental programmes is to validate computational methods, to obtain high-energy nuclear data, to characterise the performance of subcritical assemblies driven by external sources, and to develop and improve techniques for subcriticality monitoring.

With the CRP in its final year, the paper summarises, on behalf of all the participants, the status of work and preliminary CRP benchmarks results.

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\* The full paper being unavailable at the time of publication, only the abstract is included.



## EURATOM research and training programme for partitioning and transmutation

**Georges Van Goethem**

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### **Abstract**

*The EURATOM research and training programme for partitioning and transmutation is discussed. The EU has been supporting various P&T related activities since 2007 under the auspices of the 7<sup>th</sup> Framework Programme (FP7). The European Sustainable Nuclear Energy Technology Platform (SNE-TP) was put in place to help make decisions related to demonstration facilities to be built around a 2015-2020 time horizon. Two collaborative projects, ACSEPT, which studies partitioning technologies and actinide science, and EUROTRANS, which studies transmutation of high-level nuclear waste in ADS, are summarised.*

The radioactive waste which is produced in nuclear fission power systems is mainly contained in the spent fuel, which is highly radioactive for a long time. The main concern in the disposal of this waste is related to the long-lived radionuclides, some of which will remain hazardous for tens of thousands of years.

When considering sustainable energy development worldwide, one of the challenges of nuclear energy is the minimisation of the production of this long-lived radioactive waste, but also the optimisation of the use of natural resources with an increased resistance to proliferation.

Large efforts to resolve this issue are under way worldwide, concentrating on the disposal of nuclear waste in deep geological repositories. Parallel to this approach, the strategy of partitioning and transmutation (P&T) of high-level nuclear waste is investigated. P&T, associated with a multi-recycling of all transuranics (TRU), being the minor actinides (MA) and possibly plutonium (Pu), should play a key role in the development of a sustainable nuclear energy.

Any step towards the technological realisation of P&T systems could have a positive influence on the improvement of public acceptance of nuclear fission electricity production. This will enhance the actual nuclear renaissance in Europe and worldwide, which in turn would reduce Europe's steadily increasing dependency on energy imports.

Even if a final repository would still be needed for the losses of the P&T systems and for the high-level wastes which do not undergo the transmutation process, the strategy of P&T could reduce the radiotoxicity of high-level wastes and thus ease the long-term safety issue of a final repository. 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 stored in final repositories. One of the most tangible outcomes of P&T will be the reduction of the monitoring period to technological and manageable time scales.

Possible strategies can range from stable or expanding nuclear energy scenarios (with TRU treated either in dedicated transmuters in a separate fuel cycle stratum or in Generation IV fast reactor systems associated with a closed cycle) up to the scenario of a nuclear phase-out. As a consequence, in an expanding nuclear energy scenario, P&T would permit the transition from the currently practiced mono-recycling of plutonium in light water reactors (LWR) to actinide (U, Pu, MA) recycling. Even in a phase-out scenario, the combination of P&T and dedicated burners such as accelerator-driven system (ADS) technologies, but this time at regional scale, would allow meeting the above objectives of minimising the radiotoxicity, volume and heat load of high-level nuclear waste to be ultimately disposed of.

Despite the different national strategies envisaged for managing nuclear wastes, the options for P&T 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 facilities. Considering the renewed interest for closed fuel cycles in many countries, it is quite natural to strengthen the links and synergies between partitioning and 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) and, in particular, in the European Sustainable Nuclear Industrial Initiative (ESNII/fast neutron reactors and closed fuel cycle in the EU) in support of the SET-Plan [1].

To implement this strategy on the horizon 2040-2050, it is expected around 2012 to review national positions, as well as the impact of the P&T strategies on geological repository in terms of requirements and capacity. Scenarios for P&T implementation and the evaluation of technological options should also take into account the national capacities in fuel reprocessing and fuel fabrication as well as in construction of innovative reactor systems, notably during the transient period. Priorities should also be set following the review of ADS vs. critical fast systems potentialities and their different coolants. All these reviews should lead to making decisions on demonstration facilities to be built at a time horizon 2015-2020.

By joining together a great number of partners coming from European universities, nuclear research bodies and major industrial players in multi-disciplinary consortia, the FP7 Euratom Fission Collaborative Projects ACSEPT [2] and EUROTRANS [3] provide a structured R&D framework (also including non-European partners) to achieve the sound basis and fundamental improvements for future demonstrations at the pilot level.

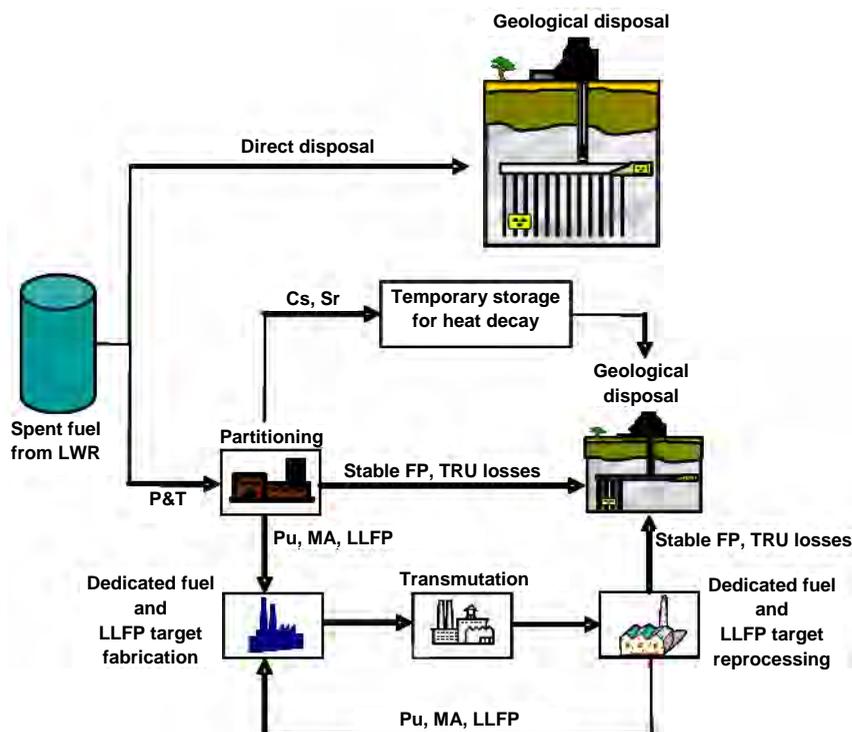
In parallel, training and education programmes are also implemented to share knowledge among the P&T community and the present and future generations of researchers. Specific attention is given to the funding of multi-disciplinary post-doctorate fellowships and to cross-cutting activities (e.g. access to large or unique infrastructures of common interest).

The implementation of P&T requires the demonstration of its feasibility at several facilities at an engineering level. At the European level, the respective research and development activities are arranged in four “building blocks” (see Figure 1):

- 1) demonstration of the capability to apply advanced reprocessing on a sizable amount of spent fuel from commercial power plants (i.e. LWR) in order to separate Pu and MA;
- 2) demonstration of the capability to fabricate at a semi-industrial level the advanced fuel needed to load a dedicated transmuter;
- 3) availability of one or more dedicated transmutors;
- 4) provision of a specific installation for processing of the dedicated fuel unloaded from the transmuter, which can be of a different type than the one used to process the original spent fuel unloaded from the commercial power plants (i.e. LWR), and fabrication of a new dedicated fuel.

**Figure 1: Implementation of partitioning and transmutation strategy**

LLFP – long-lived fission products ( $^{99}\text{Tc}$ ,  $^{129}\text{I}$ ,  $^{79}\text{Se}$ ,...), MA – minor actinides (Am, Np, Cm)



## ACSEPT project (2008-2012)

### *Partitioning technologies and actinide science: Towards pilot facilities in Europe*

The scientific and technological aspects of the above building block No. 1) is carried out in the FP7 Collaborative Project ACSEPT [2], which builds on the findings, in particular, of the FP6 project EUROPART (European Research Programme for the Partitioning of Minor Actinides and some Long-lived Fission Products from High Active Wastes Issuing from the Reprocessing of Spent Nuclear Fuels).

In line with the above timescales, the ACSEPT project is aiming at developing chemical separation processes compatible with fuel fabrication techniques, in view of their future demonstration at the pilot level.

ACSEPT is an essential contribution to the demonstration, in the long term, of the potential benefits of actinide recycling to minimise the burden on the geological repositories. ACSEPT is organised into three technical domains:

- Considering technically mature aqueous separation processes, ACSEPT optimises and selects 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.
- Concerning high-temperature pyrochemical separation processes, ACSEPT focuses first on the enhancement of the two reference cores of process selected within FP6-Europart. R&D efforts are also brought to key scientific and technical points compulsory for building a whole separation process.
- All experimental results are integrated by carrying out engineering and systems studies on hydro and pyrochemical processes 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 FP7 project FAIRFUELS.

## EUROTRANS project (2005-2010)

### *Transmutation of high-level nuclear waste in an ADS: Towards a demonstration device of industrial interest*

The scientific and technological aspects of the above building block No. 2) are carried out in the FP7 Collaborative Project EUROTRANS [3]. The objective of EUROTRANS is the design and the feasibility assessment of an industrial ADS prototype dedicated to transmutation with the following major activities:

- To carry out a first design of an experimental facility (short-term realisation) demonstrating the technical feasibility of transmutation in an accelerator-driven system (XT-ADS), as well as to accomplish a conceptual design of the European Facility for Industrial Transmutation (EFIT – long-term realisation). This step-wise approach is termed as the European Transmutation Demonstration (ETD) approach.
- To provide, for the above devices, validated experimental input (such as experimental techniques, dynamics, feedback effects, shielding, safety and licensing issues) from relevant experiments at sufficient power (20-100 kW) on the coupling of an accelerator, an external neutron source and a subcritical blanket.
- To develop and demonstrate the necessary associated technologies, especially reliable linear accelerator components, fuels, structural materials at medium to high temperature and high radiation exposure conditions, thermal-hydraulics, heavy liquid metal technologies and measurement techniques, and nuclear data.

- To prove its overall technical feasibility.
- To carry out an economic assessment of the whole system, in order to start a decision process towards a European demonstration facility.

EUROTRANS makes use of all past and foreseen developments in P&T, mainly summarised by the Thematic Network on Advanced Options for Partitioning and Transmutation (ADOPT).

The R&D work and the innovation effort to be performed within EUROTRANS are integrated into five technical domains:

- Domain DM1: DESIGN (Development of a Detailed Design of XT-ADS and a Conceptual Design of EFIT with Heavy Liquid Metal Cooling).
- Domain DM2: ECATS (Experimental Activities on the Coupling of an Accelerator, a Spallation Target and a Subcritical Blanket).
- Domain DM3: AFTRA (Advanced Fuels for Transmutation Systems).
- Domain DM4: DEMETRA (Development and Assessment of Structural Materials and Heavy Liquid Metal Technologies for Transmutation Systems).
- Domain DM5: NUDATRA (Nuclear Data for Transmutation).

The outcome of this work will allow to provide a reasonably reliable assessment of technological feasibility and a cost estimate for ADS-based transmutation, and to possibly decide on the detailed design of an experimental ADS and its construction in the future.

The next step beyond EUROTRANS should be dedicated towards the realisation of the P&T strategy: this should be done in a stepwise manner and with the ultimate goal of an industrial transmutation machine, which is the building block No. 3) mentioned above. This approach is pursued in the FP7 Project Central Design Team (CDT) for a Fast-spectrum Transmutation Experimental Facility, co-ordinated by SCK•CEN.

### Acknowledgements

The author wishes to thank the Euratom FP7 project co-ordinators (and associated consortium partners) working in the area of P&T, who generously contributed their time to produce summary reports of outstanding scientific quality for the FISA-2009 Conference [4].

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## United States programme on partitioning and transmutation\*

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### **Abstract**

*The United States Department of Energy’s Fuel Cycle R&D programme has been redirected over the past year. Rather than pursuing design and construction of current technologies for recycling components of used nuclear fuel in fast reactors, the programme is evaluating or re-evaluating all options for long-term sustainable fuel cycles. Strategies for open, modified open and fully closed fuel cycles are under development. In addition, with the cancellation of the Yucca Mountain project, the programme is evaluating options for used fuel storage, transportation and permanent disposal of high-level waste. The Fuel Cycle R&D programme will develop and demonstrate advanced technologies in order to prepare for commercial deployment of a sustainable fuel cycle by 2050.*

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\* The full paper being unavailable at the time of publication, only the abstract is included.



## Future nuclear systems: Fuel cycle options and guidelines for research\*

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### Abstract

The International Atomic Energy Agency currently estimates that some 50 countries – up from 29 today – will have nuclear reactors by 2030. If these projections are borne out, the 436 nuclear reactors currently operable around the world will be joined by more than 500 others within the next few decades. However, such a nuclear renaissance will only be accepted if the sustainability of nuclear energy is enhanced, in terms of economic, societal and environmental criteria. In particular, efficiency of the resource consumption, safety of reactor and fuel cycle facilities, acceptability and long-term safety of the waste repository as well as resistance towards proliferation are key issues to address.

In this context, nuclear systems and fuel cycles are anticipated to significantly evolve in the next decades. This paper aims to depict what are the corresponding options for the future and the corresponding supportive French R&D programme strategy, with the objective of increasing the sustainability of nuclear energy within the scope of the 1991 and 2006 French Waste Management Acts requesting in particular the study of applicable solutions for further minimising the quantity and the hazardousness of final nuclear waste.

The current French recycling strategy is based on rationale considerations:

- saving uranium resources, still at low scale, since about 10% of French nuclear electricity is produced with MOX fuels;
- mastering the growth of plutonium inventory (Pu from processing = Pu refuelled);
- producing safely and securely ultimate waste, with no plutonium in;
- keeping the plutonium concentrated in MOX spent fuels available for future use.

For the future, the long-term sustainable nuclear systems will be fast reactors which allow full use of uranium with no enrichment needs, efficient burning of plutonium and potentialities for improving waste management. They will be initially fuelled with plutonium coming from spent MOX fuels, the breeding being adjusted according to energy needs. The corresponding fuel cycle main features will be full plutonium/uranium multi-recycle, with advanced recycling concepts (no separated pure plutonium with COEXTM, on-line recycling and co-location of processing and fuel fabrication plants, appropriated international safeguards measures using a systemic approach based on accountancy and monitoring).

Regarding waste management, most of the research in France has been conducted in the framework of the 28 June 2006 Waste Management Act which defines a roadmap and the main objectives to be achieved in a near future, towards the assessment of industrial potentialities of the diverse P&T options by 2012 and the construction of the sodium-cooled ASTRID reactor

\* The full paper being unavailable at the time of publication, only the abstract is included.

*allowing minor actinide transmutation demonstration after 2020. Minor actinide recycling by P&T could significantly change the challenges for the storage of nuclear waste by reducing the heat load, the radiotoxicity and the overall “half-life” of the waste to be buried. In parallel, the retrievable geological repository will be defined in 2015 for already existing wastes, with first operation planned in 2025.*

## Minor-actinide-bearing fuel study activities in Japan for homogeneous TRU recycling fast reactor system

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### Abstract

The *Fast Reactor Cycle Technology Development Project (FaCT project)* in Japan applies a typical concept of homogenous TRU recycling with sodium-cooled fast reactors (FBR) of mixed-oxide fuel core, a simplified pelletising fuel fabrication system and an advanced aqueous reprocessing system as reference. JAEA has been promoting and conducting research and development of the FaCT project, which includes the technology developments of minor actinide separation, minor-actinide-bearing fuel fabrication, fuel property measurements and fuel irradiation. An experimental reactor (Joyo) and a prototype reactor (Monju) are the effective tools of the FaCT fuel irradiation tests. Monju restarted in 2010. The current Monju driver fuel is mixed-oxide fuel in pellet form with plutonium from LWR spent fuels. Monju will demonstrate LWR spent fuel origin plutonium utilisation in a fast reactor core as mass production driver fuel, which contains  $^{241}\text{Am}$  decayed from  $^{241}\text{Pu}$ . Post-irradiation examinations of the fuel will provide actual data to get certain knowledge of Am-bearing oxide fuel behaviour, which could be an appropriate basis for future minor-actinide-bearing fuel for the homogeneous TRU recycling concept. Some of the leading data of Am-bearing oxide fuel has been obtained from Joyo irradiation tests. For more significant utilisation of Monju core and fuel, the Global Actinide Cycle International Demonstration (GACID) project, a CEA-USDOE-JAEA collaborative activity, is in progress under the Generation IV International Forum (GIF) framework. This activity will also provide a technical basis for a future recycle demonstration programme of minor-actinide-bearing fuel in TRU homogeneous recycling strategy.

## Introduction

Japan launched the Fast Reactor Cycle Technology Development (FaCT) project in 2006 [1]. The primary concepts of future fast reactors and fuel cycle system in the FaCT project consist of sodium-cooled fast reactors, advanced aqueous reprocessing and simplified pelletising fuel fabrication. The FaCT project applies an advanced oxide fuel concept as a reference. It applies a homogeneous TRU recycling concept as one of the promising minor actinide burning concepts using fast neutrons. An advantage of the homogeneous TRU recycling concept is low minor actinide content of the fuel, which leads to a limited minor actinide contribution to fuel properties and irradiation behaviour.

In the FaCT project, the design studies and the R&D on innovative technologies regarding the main concepts are conducted with the aim to present the specific concepts of demonstration and commercial FR cycle facilities by around 2015. This activity will be followed by further developmental effort to realise a demonstration reactor around 2025 and the first commercial fast reactor before 2050. JAEA has been promoting and conducting research and development of the FaCT project, which includes the technology developments of minor actinide separation, minor-actinide-bearing fuel fabrication, fuel property measurements and fuel irradiation.

## Reference concepts of the future fast reactor and its fuel cycle [1, 2]

The reference sodium-cooled fast reactor concept of the FaCT project is the Japan Sodium-cooled Fast Reactor (JSFR, see Figure 1). The JSFR concept includes innovative technologies such as a two-loop system in the primary circuit and passive shutdown system of the reactor. For the core and fuel, a high burn-up core with ODS cladding and minor-actinide-bearing oxide fuel are proposed.

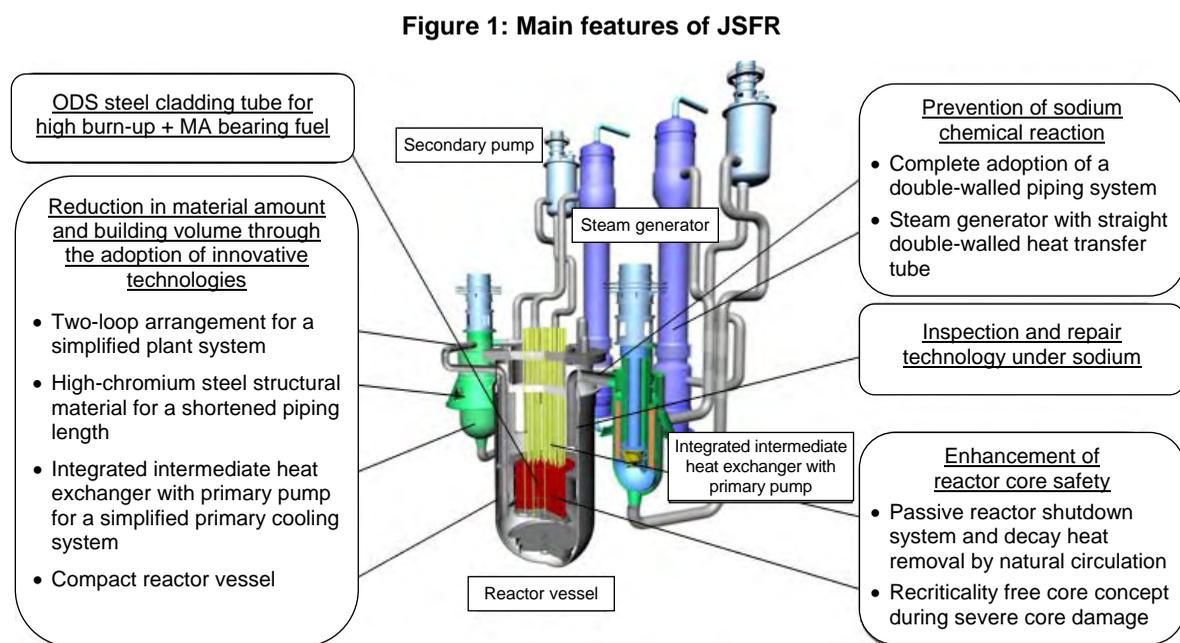
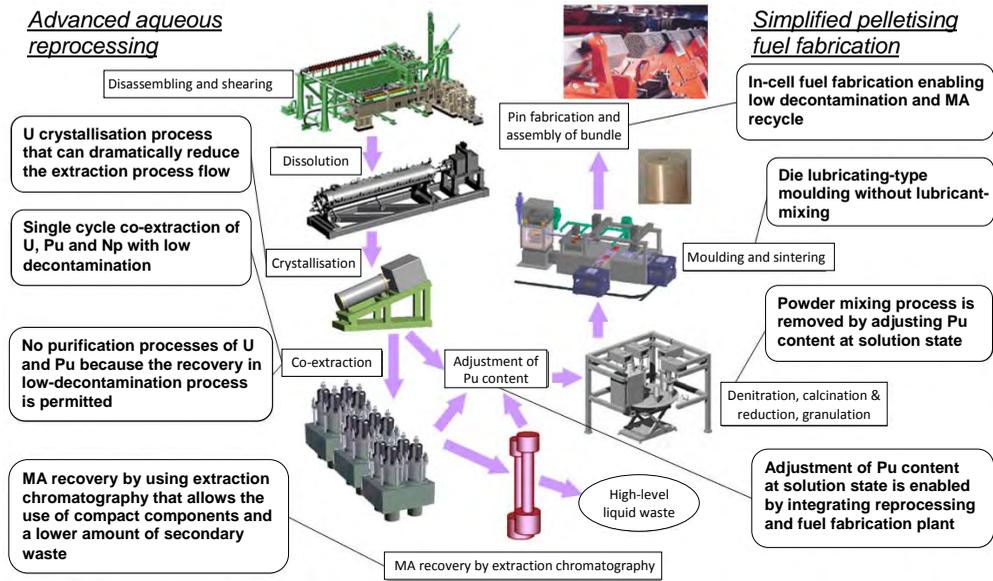


Figure 2 shows the reference concepts of reprocessing and fuel fabrication. Major features are homogeneous recycling of minor actinides and a simplified pelletising process. The simplified pelletising process is to reduce the fabrication process steps by supplying starting oxide fuel powder with adjusted enrichment and by adopting the pellet pressing method of binderless granulated oxide. This process is expected to reduce the cost and waste of fabrication and is applicable to both minor-actinide-bearing oxide fuel and the (U,Pu) oxide fuel without intended minor actinide.

**Figure 2: Main features of the advanced fuel cycle system of the FaCT project**



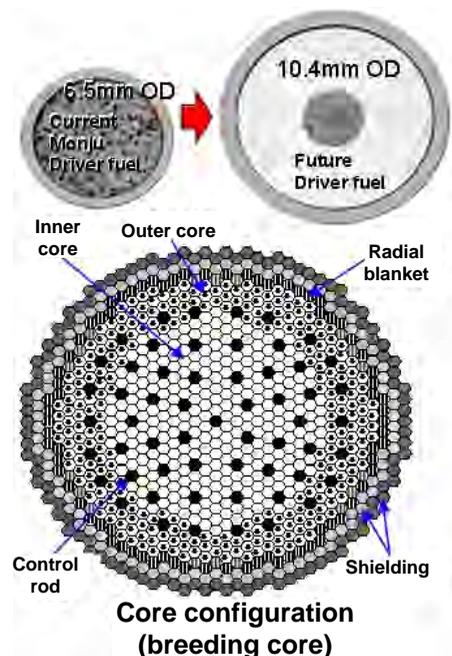
**Fuel of the FaCT project [3, 4]**

The JSFR core and fuel are designed to achieve high burn-up and high core outlet temperature, and to be capable of minor-actinide-bearing fuel loading with homogeneous recycling manner, not only for the fast reactor recycle, but also for fuel supply from LWR spent fuel. Its reference fuel is oxide fuel and minor actinide content up to 5% in heavy metal is considered.

Figure 3 shows a reference concept of JSFR core and fuel. The core outlet temperature is 550°C.

**Figure 3: Large-scale JSFR core and fuel concept**

Items	Breeding core	Break even core
Nominal full power (MWe/MWt)	1 500/3 570	←
Coolant temperature [outlet/inlet] (°C)	550/395	←
Primary coolant flow (kg/s)	18 200	←
Core height (cm)	100	←
Axial blanket thickness [upper/lower] (cm)	20/20	15/20
Number of fuel assemblies [core/radial blanket]	562/96	562/-
Envelope diameter of radial shielding (m)	6.8	←
Fuel cladding outer diameter (mm)	10.4	←
Fuel cladding thickness (mm)	0.71	←
Number of fuel pins per assembly	255	←
Wrapper tube outer flat-flat width (mm)	201.6	←
Wrapper tube thickness (mm)	5.0	←



Core fuel column length is 100 cm. Fuel pin design is large diameter, as large as about 10 mm, and high density annular pellet fuel to have low smeared density as 82%TD to achieve high BU with a high density pellet, which is a typical change from current Monju driver fuel pin which is 6.5 mm in diameter and applies low density solid fuel. Such fuel pin design provides the advantage of high fuel volume fraction in the core to achieve superior core neutronic characteristics, low fuel smeared density to accommodate fuel swelling at high burn-up and fuel fabrication economics consistent with low fuel smeared density design. We also apply a low O/M fuel to reduce the cladding inner surface corrosion at high BU.

Aiming at achieving high burn-up as 150 GWd/t of average BU, which corresponds to 250 GWd/t of peak BU and 250 dpa of fast neutron dose, oxide dispersion ferritic steel (ODS) cladding and PNC-FMS sub-assembly duct were selected as reference core materials. Using ODS cladding tube with high strength in high temperature, a design with 700°C of maximum cladding temperature is feasible.

Fuel developmental activities are in progress to realise JSFR driver fuel for the FaCT project. Irradiation tests are important activities of the development. Major irradiation tests to be performed in Joyo and in other reactors are listed below:

- ODS irradiation (material, fuel pin, fuel pin bundle);
- PNC-FMS irradiation (material, fuel pin, SA duct);
- large diameter fuel pin;
- simplified process fuel pellets;
- annular fuel PTM (PTM = power-to-melt);
- irradiated fuel PTM;
- MA-bearing oxide fuel (Am,Np-bearing, Am+Np+Cm-bearing);
- transient tests (reactor tests and hot cell tests, burn-up extension of current fuels).

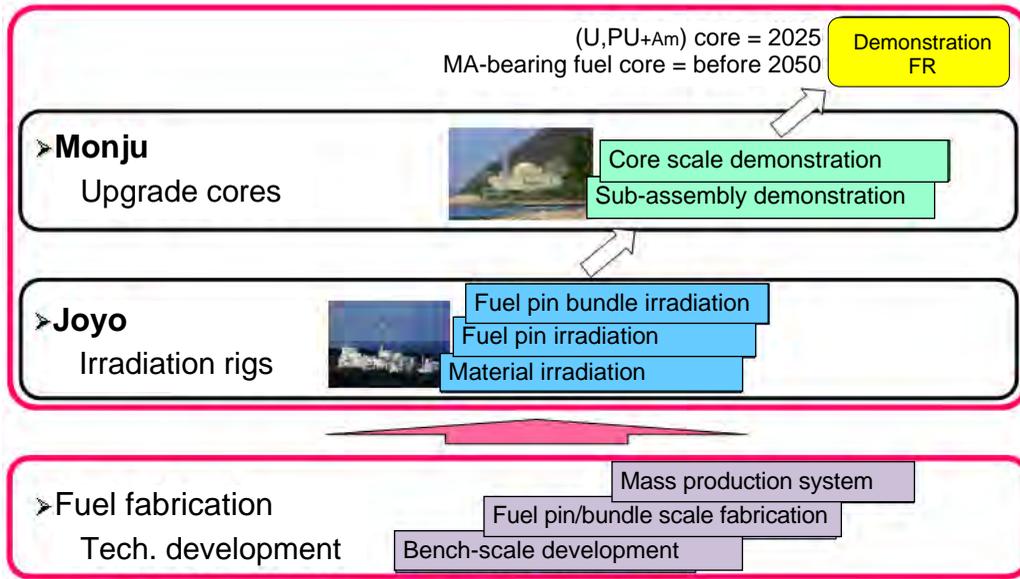
Fuel fabrication technology is one of the key issues of future fast reactor fuel. The simplified pelletising process under development aims at reducing the pellet fabrication processes due to less oxide fuel powder treatment processes and less organic additives [2]. Its key technologies are homogeneous oxide powder supply by the microwave conversion process of U-Pu solution, binderless granulation and die-lubrication pressing. The microwave conversion process was already established and development of the other two technologies is making good progress. Irradiation behaviour of fuel pellets made from microwave conversion powder was preliminarily investigated as well.

Figure 4 shows a basic scheme of fuel development including irradiation tests and fuel fabrication technology development. Using Joyo irradiation rigs, material irradiation tests, fuel pin irradiation tests and fuel pin bundle irradiation tests will be performed. Based on those irradiation experiences and test data, SA demonstration irradiation and core scale demonstration will be undertaken in Monju upgrade cores. These are the leading irradiation tests to realise demonstration reactor cores, which is expected to start at around 2025.

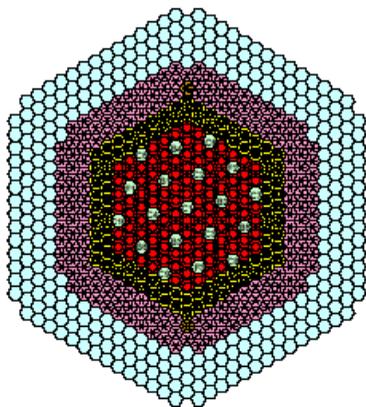
### Monju and its fuel with Am [5-7]

Monju is a prototype sodium-cooled fast reactor with 714 MWth and 280 MWe. It has a loop-type reactor system with three primary and secondary loops. Reactor vessel inlet and outlet temperatures are 397°C and 529°C, respectively. Figure 5 shows the major characteristics of the current Monju core. The core is a homogeneous two-region core of oxide fuel with axial and radial blankets. The core fuel column length is 930 mm and core equivalent diameter is around 1 800 mm. The driver fuel pin diameter is 6.5 mm and the fuel pellet is solid, with 85% theoretical density.

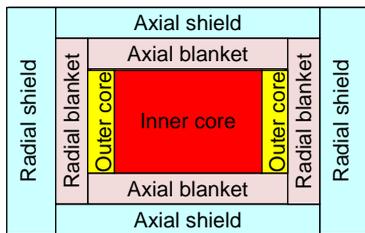
**Figure 4: Basic scheme of fuel development of FaCT project**



**Figure 5: Major characteristics of current Monju core**



Horizontal cross-section



Vertical cross-section

Core zone	Inner core		108
	Outer core		90
Radial blanket			172
Control rod	Fine control rod (FCR)		3
	Coarse control rod (CCR)		10
	Back-up control rod (BCR)		6
Neutron source			2
Neutron shield			324

Principal design and performance data of Monju

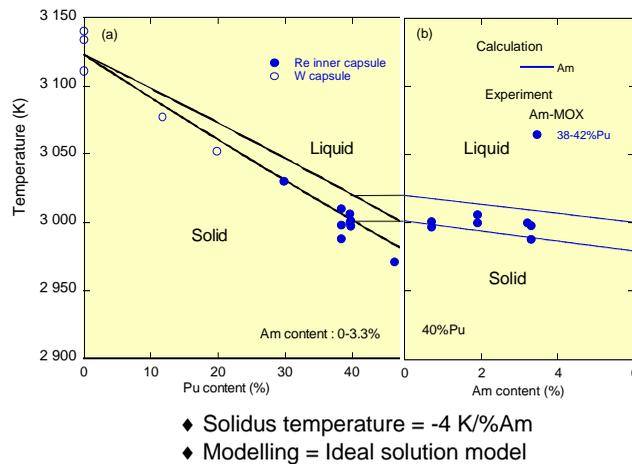
Reactor type	Sodium-cooled loop-type
Thermal output	714 MW
Electrical output	280 MW
Fuel material	PuO <sub>2</sub> -UO <sub>2</sub>
Core dimension	
Equivalent diameter/height	1.8/0.93 m
Blanket thickness	
Upper/lower/radial equivalent	0.3/0.35/0.3 m
Plutonium fissile enrichment	(Inner core/outer core)
Fuel of initial core Type 1	15/20 wt.%
(SST core) Type 2	16/21 wt.%
Type 3	16/21 wt.%
Fuel of equilibrium core	16/21 wt.%
Fuel inventory	
Core (U+Pu+Am-241 metal)	5.9 tonne
Blanket (U metal)	17.5 tonne
Average burn-up	
Equilibrium core	80 000 MWd/t
Cladding material	SUS316
Cladding outer (diameter/thickness)	6.5/0.47 mm

Monju restarted in May 2010 after 14 years and five months suspension. During the suspension, <sup>241</sup>Am accumulated due to the decay of <sup>241</sup>Pu, which has a half-life of about 14 years. The Monju restart core contained about 1.5% of <sup>241</sup>Am in the whole core average [6, 7].

### Am-bearing fuel study [8-10]

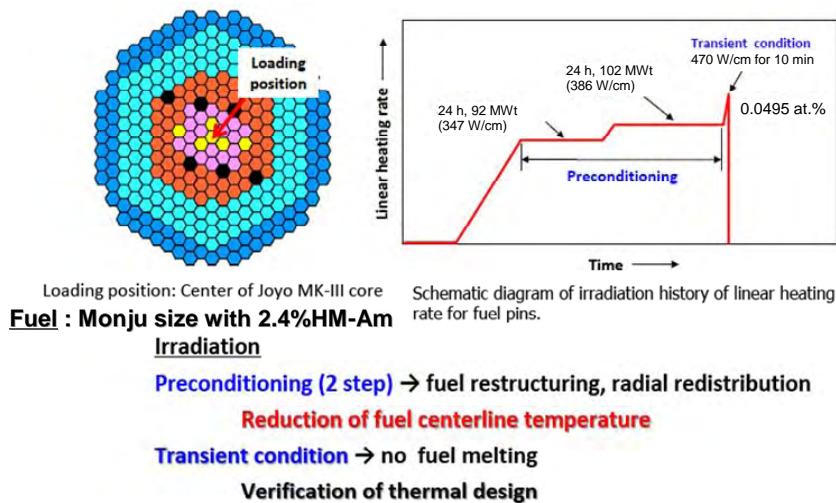
Aiming at the evaluation of Am-bearing oxide fuel, fuel property studies and fuel irradiation tests were performed. Fuel property study is a significant issue of MA-bearing oxide fuel. JAEA has been investigating MA-bearing fuel properties using out-of-pile experimental studies and analytical studies [8]. Major properties such as fuel melting temperature, thermal diffusivity, specific heat and oxygen potential have been experimentally studied. Figure 6 shows typical results of fuel melting temperature measurements. Current results show the limited contribution of minor actinides on fuel properties for oxide fuels of homogenous recycling of minor actinides.

**Figure 6: Melting temperature of Am-bearing oxide fuel**



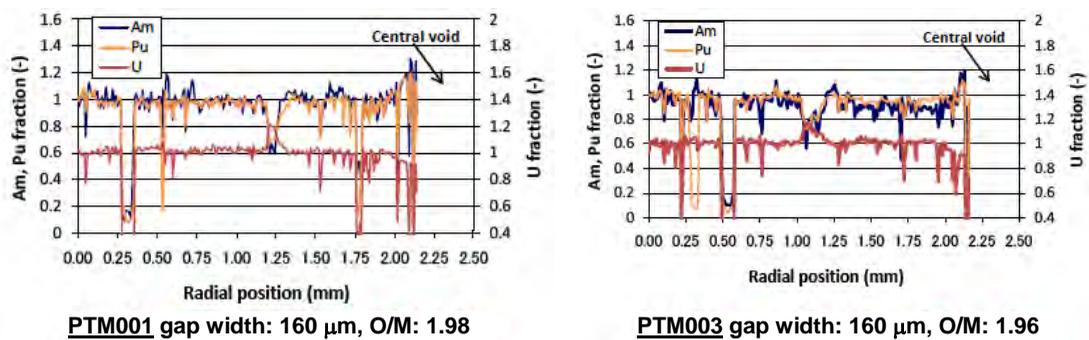
Am-bearing oxide fuel with Monju fuel pin specifications were irradiated in Joyo [9]. The Am content of the test fuel reached 2.4% to cover the Am content of Monju fuel. Irradiation conditions were selected to simulate the Monju power rise at the beginning of the operation cycle and to cover the overpower at the upset condition in Monju core design. Fuel pin linear heat rate reached about 470 W/cm and was maintained for 10 minutes. Figure 7 shows its power history in Joyo irradiation.

**Figure 7: Power history of Am-MOX fuel irradiation in Joyo**



Short-term irradiation testing of MA-bearing oxide fuel supplied important irradiation data such as early-in-life fuel restructuring of MA-bearing fuel and MA redistribution as well as Pu redistribution. It is very important to be aware of the MA redistribution behaviour at the beginning of life, since it may limit the maximum LHR of MA-bearing fuel due to the melting limit of licensing design evaluation. After irradiation, fuel microstructures were investigated and confirmed that fuel restructuring behaviour is identical with (U,Pu) fuel without MA, and that fuel maximum temperature of Am-bearing oxide fuel with 2.4% Am content was below the melting point. Am and Pu redistribution data were also obtained during the post-irradiation examination as indicated in Figure 8. They showed that low O/M fuel reveals less redistribution than the high O/M fuel. Other MA-bearing oxide fuel such as Am,Np-bearing fuel were irradiated in Joyo up to 10 minutes and 24 hours at maximum power [10]. These irradiation test data and post-irradiation examination data of Monju Am-bearing fuel will provide actual data to further knowledge of Am-bearing oxide fuel behaviour to contribute to quantitative analyses of future driver fuel design.

**Figure 8: Early-in-life radial redistribution of actinides**



### Further development of homogeneous recycling TRU oxide fuel

Further developmental effort will be made, which shall include fuel property study and fuel pin bundle irradiation testing, as well as fuel pin irradiation tests such as power-to-melt and fuel fabrication technology development.

International collaborations are indispensable to promote fast reactor fuel development. Generation IV International Forum (GIF) is a relevant framework of such collaborations. One such collaboration is the Global Actinide Cycle International Demonstration (GACID) which includes CEA France, USDOE and JAEA Japan, under the GIF sodium-cooled fast reactor (SFR) system arrangement [11]. The GACID project aims at demonstrating MA transmutation capability and MA-bearing fuel integrity in a fast reactor core, using Joyo and Monju. The project consists of three steps as listed below and a series of irradiation tests in Joyo and Monju:

- 1) *Step 1: Precedent Limited MA-bearing Fuel Preparatory Irradiation Test.* This test assumes  $^{237}\text{Np}$  and  $^{241}\text{Am}$  only as for the MA. Moreover only a single pin-scale irradiation test in Monju is planned. Therefore this test is expected to be implemented at an earlier stage of the project. Figure 9 shows its scheme.
- 2) *Step 2: Pin-scale Cm-bearing Fuel Irradiation Test.* A full range of MA composition is assumed for this test. Not only Np and Am but also Cm will be contained in the test fuel, although the test will be conducted on a pin scale. A precedent irradiation test in Joyo is being planned for Monju irradiation licensing.
- 3) *Step 3: Bundle-scale MA-bearing Fuel Irradiation Demonstration.* After completing the above two steps of the precedent irradiation tests, the final goal, bundle-scale full-range MA-bearing fuel irradiation demonstration, will be performed in Monju. This technical demonstration will be undertaken in a reasonable time frame, and the whole project is to be conducted over a period of 20 years.

Figure 9: Schematic view of the GACID Step 1 Test

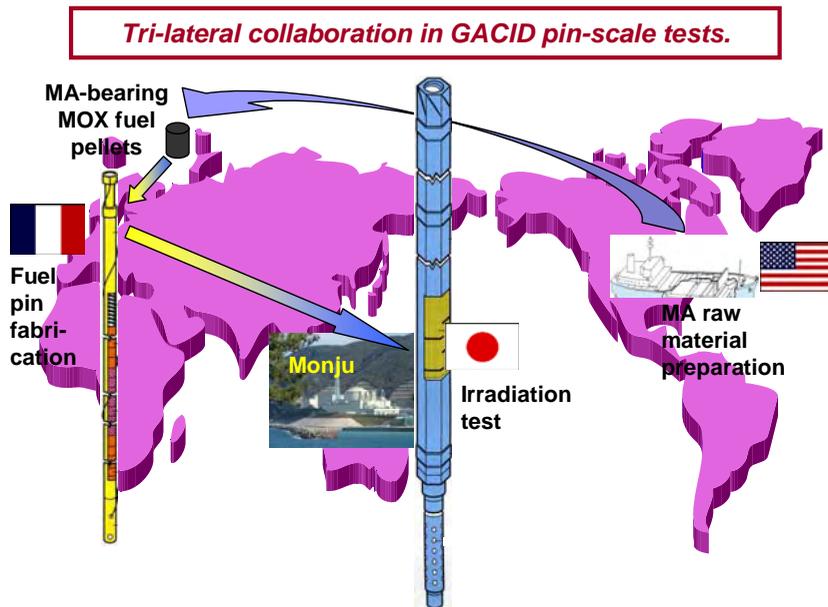
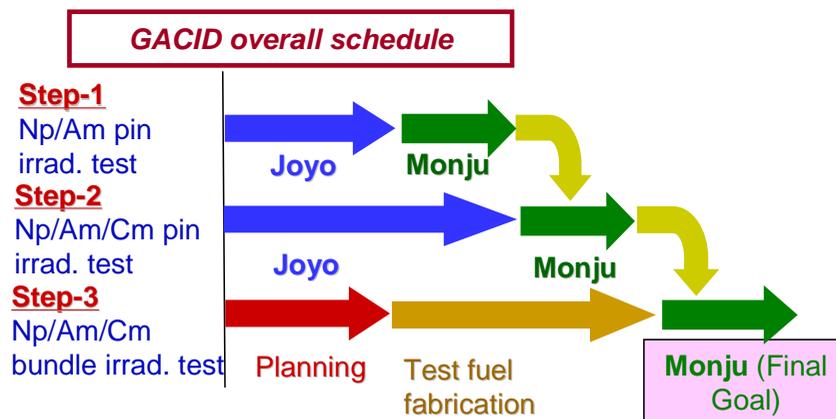


Figure 10 shows the overall plan of the GACID project. The project is in progress and the following activities are underway in each participating organisation:

- MA raw material preparation and shipping;
- MA-bearing MOX fuel pellet sintering;
- material property measurement;
- precedent Joyo irradiations and PIE;
- licensing in Monju and Joyo;
- preliminary programme planning for bundle-scale irradiation demonstration.

The GACID project is a good example of Monju utilisation for international collaboration, which accelerates the developmental programme and will lead to more efficient outcomes.

Figure 10: Overall plan of the GACID project



## Conclusions

Fuel development for future fast reactors is in progress as part of the Japanese FaCT project. The reference fuel of the FaCT project is MA-bearing oxide fuel in homogeneous recycling. Developmental efforts include irradiation tests, fuel fabrication technology development and out-of-pile studies such as fuel property investigations.

Monju restarted its core with Am-bearing fuel and its characteristics were experimentally evaluated. Further experiences and examinations of minor-actinide-bearing fuel are expected in the future Monju core. Monju core and fuel are expected to play an important role, as is Joyo, to demonstrate the technical feasibility of TRU homogeneous recycling fuels.

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## Perspectives of pyroprocesses for long-term spent fuel management in Korea

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### Abstract

*Korea has 20 nuclear power reactors, generating about 700 t/yr of spent fuel. The spent fuel pools at some of its power reactors will be full by 2016. One promising option for spent fuel management is the treatment of spent fuel by pyroprocessing and the recycling of recovered TRU to the transmutation reactor. The Korea Atomic Research Institute (KAERI) has been developing pyroprocessing technology, which comprises pretreatment, electroreduction, electrorefining, electrowinning and a waste salt treatment system. The basic concept of pyroprocessing is group recovery, which enhances the proliferation resistance significantly, as sole plutonium cannot be separated. A part of the uranium recovered from spent fuel is blended with this plutonium and minor actinide mixture, and the blended material is fabricated as the fuel of a sodium-cooled fast reactor (SFR). The plutonium and minor actinide mixture is burnt out in the SFR, resulting in the transmutation of the long half-life nuclides. This reduces the waste management period by 1/1 000 from 300 000 years.*

## Introduction

Nuclear energy has recently received more attention as a green energy source, since the emissions of greenhouse gases such as carbon dioxide into the environment are reduced. Based on this environmentally friendly aspect as well as on energy demand, numerous nuclear power plants, including 12 additional plants in Korea, are under construction or being planned worldwide. However, an important intrinsic issue must still be resolved, i.e. spent fuel management. Spent fuel will increase with the number of nuclear power plants. A recent projection puts spent fuel accumulation at 445 000 tHM in 2020 [1]. As of the end of 2008, an accumulation of spent fuel is more than 10 000 tHM in Korea.

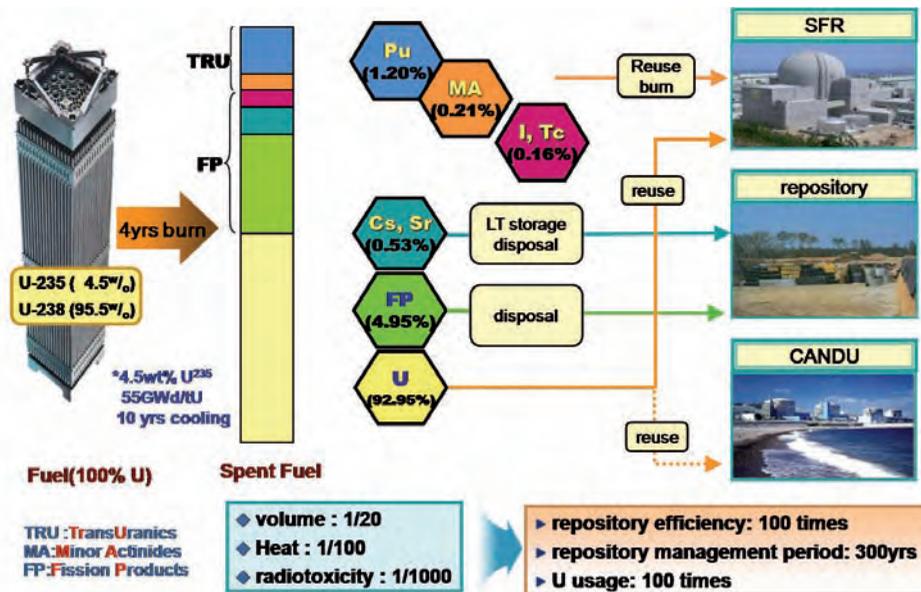
The treatment of spent fuel can be processed by two major methods, the aqueous process and the pyroprocess. PUREX is a typical aqueous process that uses an aqueous solution to treat spent fuel. A pyroprocess uses molten salt instead of an aqueous solution. Aqueous processes are more proliferative than the pyroprocess, as the former is able to separate pure Pu, whereas the latter recovers Pu together with other impurities, such as MA.

## KAERI's pyroprocessing technology

The basic concept of pyroprocessing is group recovery, which enhances proliferation resistance significantly, as sole Pu cannot be separated. Pu with minor actinides, which comprise 1.4% of spent fuel, is recovered as a group. A part of uranium recovered from spent fuel is blended with this Pu and MA mixture, and the blended material is fabricated as the fuel of a SFR. The Pu and MA mixture is burnt out in the SFR, resulting in the transmutation of the long half-life nuclides. This reduces the waste management period by 1/1 000 from 300 000 years.

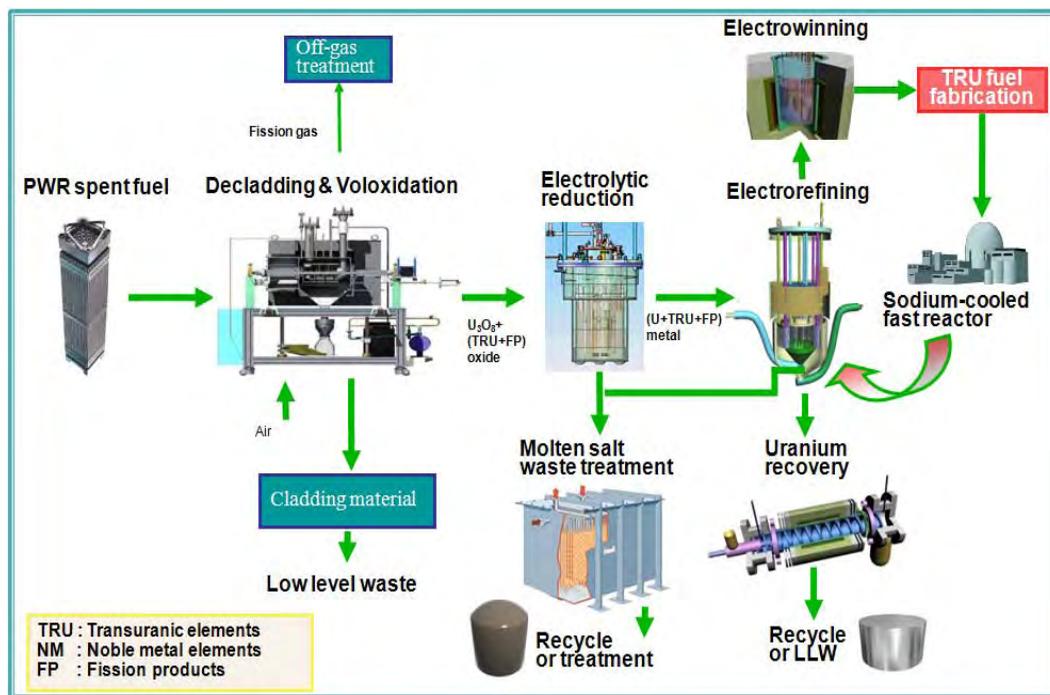
Cs and Sr, which are major components regarding heat load in spent fuel, are recovered during pyroprocessing, stored separately, and subsequently disposed of. The removal of TRU, Cs and Sr from spent fuel allows the repository burden to be reduced by 1/100, compared to a case where there is no removal. The fission products (FP) are recovered and transferred to a repository. As a result of pyroprocessing, both the repository efficiency and uranium usage increased up to 100-fold, as shown in Figure 1.

Figure 1: Pyroprocess treatment concept of spent fuel



The KAERI pyroprocess, as shown in Figure 2, includes disassembling, chopping and decladding of a spent fuel, a voloxidation to remove the volatile fission products, an electroreduction to reduce oxide spent fuel to metal, an electrorefining to recover pure uranium, an electrowinning to recover the TRU mixture and waste salt treatment to minimise the waste salt generation and the fabrication of a final waste form. In terms of high capacity and high throughput these pyroprocessing technologies have been significantly improved recently.

Figure 2: Schematic illustration of the KAERI pyroprocess



In the electrolytic reduction process, the spent fuel oxide material from voloxidation is prepared for use in the electrorefiner by reducing the oxide to a metal [2, 3]. The electrorefining process cannot accept oxide nuclear fuels directly, because the actinide oxides do not dissolve into the chloride salt. Also, in order to be used in an electrorefiner, materials must be electrical conductors. The metallic form produced by the electrolytic reduction process is suitable for use in the electrorefining process. In the electrolytic reducer, the feed material is placed in a fuel basket. A voltage is applied between the fuel basket cathode and a platinum anode. This process uses a molten  $LiCl$  salt electrolyte with a small amount of  $Li_2O$  in solution at  $650^\circ C$ . Spent fuel metals are produced in the cathode basket and oxygen is evolved from the anode. Fission products, such as Cs, Sr and Ba, are dissolved into the salt in this process, forming chloride compounds in the electrolyte salt. The metal that is collected in the cathode basket contains all of the actinides, lanthanides and metallic fission products present in the spent fuel. After the electrolytic reduction process, the residual salt material is ~20 wt.% of the cathode material. The cathode process for the removal of residual salts in the cathode basket of the electrolytic reducer can be added to ease the burden of the salt management process. The resulting metal is suitable for direct introduction into the electrorefiner.

A high throughput electrorefining system (HTER), which has the purpose of separating pure uranium from the impure electrolytically reduced spent fuel, is composed of an electrorefiner, a salt distiller and a melting furnace. A U-chlorinator ( $UCl_3$  making equipment) and a transportation system are also needed to operate the HTER. In the electrorefiner, uranium deposition is initiated in molten  $LiCl-KCl$  salt with about 9 wt.%  $UCl_3$ . Uranium dendrites are then deposited and fall from the electrode spontaneously [4], and are finally collected at the bottom of the reactor. The collected uranium deposits are transferred with a screw to a

container. The uranium deposits are fed into a salt distiller, and salt in the uranium deposits is distilled by using a salt distiller. The salt-distilled uranium is melted and subsequently reformed to ingots for fabrication to nuclear fuel for SFR.

Electrowinning technology is implemented to recover TRU from the molten salt system, a major process in the pyroprocessing technology, with proliferation resistance. The electrowinning technology of LCC is employed to recover group actinides such as uranium and TRU (Np, Pu, Am, Cm) in the molten salt (LiCl-KCl), which is transferred from the electrorefining process. A cadmium distillation technology is also used to separate Cd and actinides from recovered actinide/Cd products by LCC, while the residual actinide recovery (RAR) is incorporated for the treatment of spent salt with low concentrations of actinides.

During the pyroprocessing of LWR spent oxide fuels, two different waste salts are generated: i) LiCl waste containing alkali and alkaline-earth (Group I/II) fission products (FP) from the electrolytic reduction process; ii) LiCl-KCl eutectic salt waste containing rare-earth FP from the electrowinning (RAR) process. Since these waste salts are radioactive, heat-generative and highly soluble in water, they must be fabricated into durable waste forms that are compatible with the environment inside a geologic repository for a long time [5]. KAERI is working on two key R&D concepts for the development of innovative waste salt treatment technologies: i) the minimisation of waste salt generation by the removal of fission products in the waste and then recycling of the cleaned salt to the main processes; ii) the increase of safety by a fabrication of high-integrity final waste form. KAERI has accordingly been developing various FP removal and waste solidification technologies such as melt crystallisation, oxidative precipitation and SAP/ZIT solidification.

Each unit process has been tested in lab-scale, and these unit processes will be incorporated in the PRIDE (Pyroprocess Integrated Inactive Demonstration) facility in order to evaluate the scale-up design on the basis of the information from lab-scale experiments, the integrity of each unit performance, safeguards of the integrated process, etc. Moreover, the demonstration of an engineering-scale (10 tHM/yr) pyroprocessing system is scheduled for 2016.

In parallel with pyroprocessing technology, the SFR is being developed in KAERI and a demonstration SFR is planned to be constructed by 2028. TRU and uranium extracted through pyroprocessing will be used to fabricate metallic fuel, U-TRU-Zr for SFR. Currently fabrication technology and the qualification of metallic fuel are being investigated.

A geological disposal system for HLW from pyroprocess has been developed. The copper-cast iron overpack is designed to withstand for more than 1 000 years. According to the thermal analysis on the basis of present waste salt treatment, the spacings between the tunnels and between the boreholes are 40 m and 5 m, respectively, and the disposal area reduces to less than 1/70 compared with the direct disposal of spent fuels.

## Summary

The pyroprocess R&D in KAERI aims to increase equipment throughput and to reduce the volume of final waste to be disposed of. The application of innovative technologies such as the adoption of a graphite cathode in the electrorefiner and waste salt regeneration by a crystallisation method was tested. Based on the results of bench- and laboratory-scale tests, an inactive engineering-scale integrated pyroprocess (PRIDE) facility is planned to be constructed and operated by the end of 2016. Active tests in an engineering-scale pyroprocessing facility (ESPF) will follow.

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## **Session I**

### **Fuel Cycle Strategies and Transition Scenarios**

***Chairs: T. Mizuno, B. Dixon***



## International comparison for transition scenario codes involving COSI, DESAE, EVOLCODE, FAMILY and VISION

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### Abstract

Several scenario codes have been developed to study the future of nuclear energy in different countries and institutes. These codes allow simulating scenarios for nuclear energy at the national, regional or worldwide level. They simulate the whole fleet of nuclear reactors with associated fuel cycle plants and storage for nuclear materials. The codes enable to take into account the dynamic transition between an initial and a final situation for nuclear energy. The results are mainly the flux and the composition of heavy elements depending on time: natural uranium needs, enrichment needs, fresh fuel fabrication needs, fuel irradiation, inventory of spent fuel and nuclear materials, reprocessing needs.

The Expert Group of Fuel Cycle Transition Scenario (FCTS) is working under the guidance of the Working Party on Scientific Issues of the Fuel Cycle (WPFC) of the OECD Nuclear Energy Agency (NEA). National, regional or worldwide transition scenarios are studied inside this expert group with different existing tools devoted to scenario studies. After a review on existing national scenarios, one of the first missions of this expert group is to compare the existing scenario codes in terms of capabilities, modelling and results. Thus, it was decided to perform a benchmark between the existing codes.

## Purpose of the benchmark

Five scenarios codes were involved: COSI 6 developed at CEA (France), DESAE 2.2 developed at ROSATOM (Russia), EVOLCODE developed at CIEMAT (Spain), FAMILY 21 developed by JAEA (Japan) and VISION 2.2 developed at INL (United States). The purpose was to compare the methodologies used by the codes and a set of important results. The benchmark is based on the version of the codes available at the time and newer versions may have added capabilities.

Three different scenarios were selected having three different level of complexity. The first scenario is the open cycle simulating a LWR fleet with the direct disposal of spent fuel. The second scenario is more complex and simulates the single recycling of plutonium in the LWR. The third scenario is the most complex and simulates the transition between a LWR fleet and a Generation IV fast reactor fleet recycling Pu and minor actinides.

## Presentation of the codes

The possibilities offered by the codes are described in Table 1. The table indicates that these codes have different capabilities, particularly concerning the physical models. Thus, to make the comparison possible, only the common capabilities were compared and the benchmark focused on fuel and heavy nuclide calculations: U, Pu, Am, Np and Cm.

**Table 1: Comparison of code capabilities**

	COSI 6	DESAE 2.2	EVOLCODE	FAMILY 21	VISION 2.2
Language/software	Java		Fortran	Microsoft Visual Basic (Japanese edition)	System Dynamics/Power Sim
Facilities: discrete/continuous	Discrete	Continuous	Discrete	Discrete	Continuous
Fuel (batches)/continuous	Discrete	Discrete	Discrete	Discrete	Continuous (except for ordering of first cores)
User interface	Graphical user interface	Graphical user interface	Text interface	Graphical user interface and Microsoft Excel	MS-Excel spreadsheet and/or graphic user interface
Simultaneous advanced technologies scenarios	Any combination of LWR, HTR, FR (SFR and GFR), ADS + different types of fuels	Yes	Any reactor with any fuel	Any combination of LWR, HWR, FR (SFR, GFR, LFR and ADS + different types of fuels)	One-tier, two-tier scenarios (+ choice of the number of recyclings)
Isotopic tracking	Y (isotopes of U/Pu/MA/200 FP)	U, Pu, minor actinides	Yes (~3 300 isotopes)	Yes (isotopes of U/Pu/MA/880 FP)	Yes (follows up to 81 isotopes)
Choice of fuel	User	User	User	User	User
Calculation of transmutation performance in cores	Embedded CESAR code (Fortran) with one-group cross-section libraries based on deterministic APOLLO 2 and ERANOS systems; direct coupling with ERANOS possible	No coupling with transmutation code	Creation of one-group cross-sections with EVOLCODE 2; possibility of choosing reference libraries.	Stored depletion matrix based on results of depletion calculation by the ORIGEN2 code	Pre-calculated fuel recipes with interpolation (as a function of the number of cycles)
Start-up and shutdown fuel loads	Yes	Start-up only	Yes	Yes	No

**Table 1: Comparison of code capabilities (cont.)**

	<b>COSI 6</b>	<b>DESAE 2.2</b>	<b>EVOLCODE</b>	<b>FAMILY 21</b>	<b>VISION 2.2</b>
Front-end fuel cycle facilities	All facilities represented	Enrichment	Enrichment	Enrichment Fabrication	Only fuel fabrication and enrichment facilities represented
Reprocessing plants	Represented	Represented	Represented	Yes	Represented
Reprocessing capacity deployed	Automatic/ manual	Manual	Yes	Automatic/ manual	Automatic/ manual
Spent fuel to be reprocessed	User choice: "first-in first-out" or "last-in first-out"	First-in first only	User choice: "first-in first-out" or homogeneous	User choice: "first-in first-out" or "last-in first-out"	User choice: oldest/youngest (with min. cooling time) and from on-site/MRS/ retrievable repository
Fissile material availability forecast	No	No	No	No	Y (user-defined degree of conservatism in ordering new NPP)
User parameter for deployment of reactors	No	No	Yes	Yes	User-defined scenario/growth driven/level of conservativeness
Waste – radioactivity	Yes	No	Yes	Yes (at vitrified waste production step)	Yes
Waste – decay heat	Yes	Yes	Yes	Yes (same as above)	Yes
Waste – radiotoxicity	Yes	Yes	Yes	No	Yes
Waste conditioning modelling	Yes	No	No	Yes (heat rate and weight of oxides are considered at vitrified waste production)	Yes
Repository requirement assessment	Yes	No	No	No	Yes (several types of disposal)
LLW modelling	Yes	No	No	No	Yes (A/B/C/GTCC)
Economics assessment module	Yes	Yes	No	No	Yes
Economics optimisation	No	Yes	No	No	Uncertainty analysis
U price model	No	Yes	No	No	Uncertainty range
Transportation costs	Yes	Yes	No	No	No
Proliferation metrics	Heating rate from Pu (W/kg)	No	No	Heating rate from Pu (W/kg) at Pu storage and MOX fuel fabrication steps	Yes – multiple
	Weight fraction of even isotopes	No	No	Weight fraction of U, Pu, MA isotopes	
	Inventory (significant quantities defined by IAEA)	No	No	Amount of Pu storage after reprocessing	
	Concentration (SQ/t)	No	No		

### Scenario assumptions

Scenario 1 simulates an open cycle nuclear fleet, Scenario 2 simulates the monorecycling of Pu in the PWR and Scenario 3 simulates the continuous recycling of Pu and MA in the fast reactors. The main reactor and fuel cycle assumptions are listed in Table 2.

Figure 1: Scenario 1 – flow chart and installed capacity

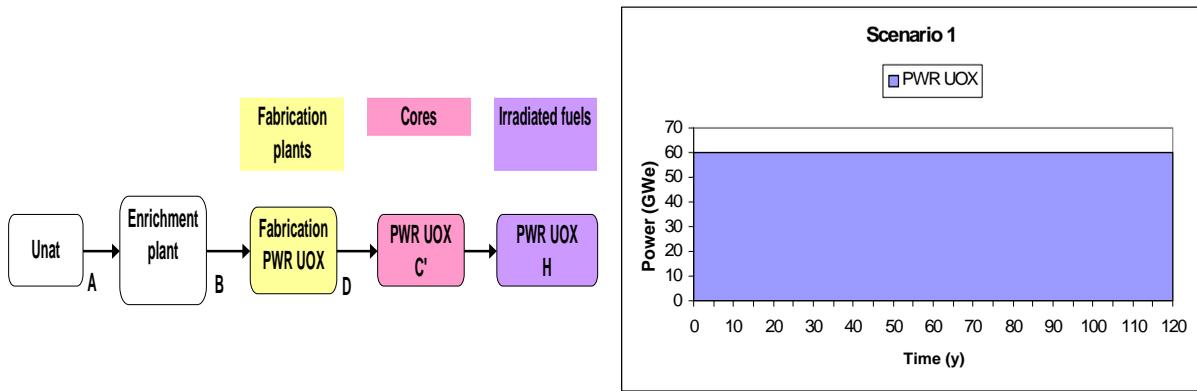


Figure 2: Scenario 2 – flow chart and installed capacity

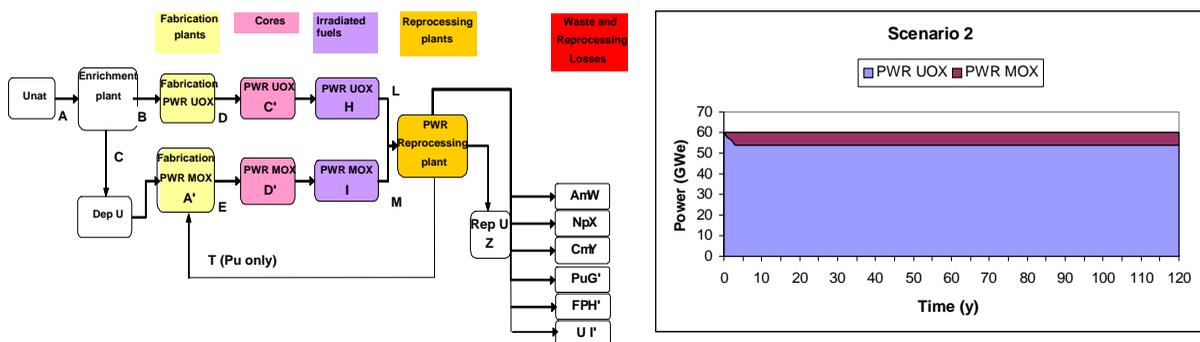
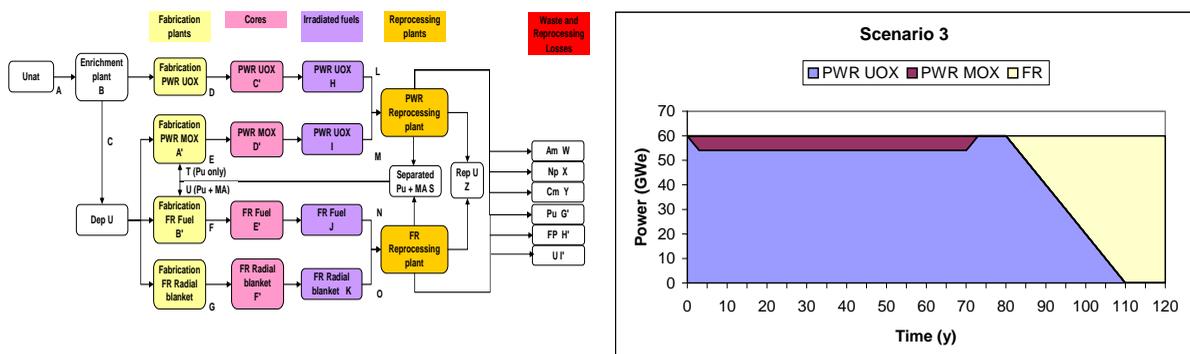


Figure 3: Scenario 3 – flow chart and installed capacity



**Table 2: Data compilation for the benchmark study**

		PWR UOX	PWR MOX	FR
<b>Fuels/blankets</b>				
Fissile burn-up	GWd/tHM	60	60	136
Axial blankets burn-up	GWd/tHM	–	–	15
Radial blankets burn-up	GWd/tHM	–	–	25
Minimum cooling time	y	5	5	2
Fabrication time	y	2	2	2
Fresh fuel <sup>235</sup> U enrichment	%	4.95	0.25	0.25
Moderation ratio		2	2	–
Equivalent Pu content	%	–	–	14.5
<b>Cores</b>				
Electrical nominal power	GW	1.5	1.5	1.45
Efficiency	%	34	34	40
Load factor	–	0.8176	0.8176	0.8176
Heavy metal masses				
Fissile	t	128.9	128.9	41.4
Axial blanket	t	–	–	18.0
Radial blanket	t	–	–	13.5
Breeding gain		–	–	≈1
Cycle length	EFPD	410	410	340
Core fraction (fuel)		1/4	1/4	1/5
Core fraction (radial blankets)		–	–	1/8
<b>Reprocessing plants</b>				
Priorities		First-in first-out	First-in first-out	First-in first-out; first fuel then blankets
Losses (U and Pu)	%	0.1	0.1	0.1
<b>Initial spent fuels</b>				
Initial mass	t	10 000	0	0

## Results

### Scenario 1

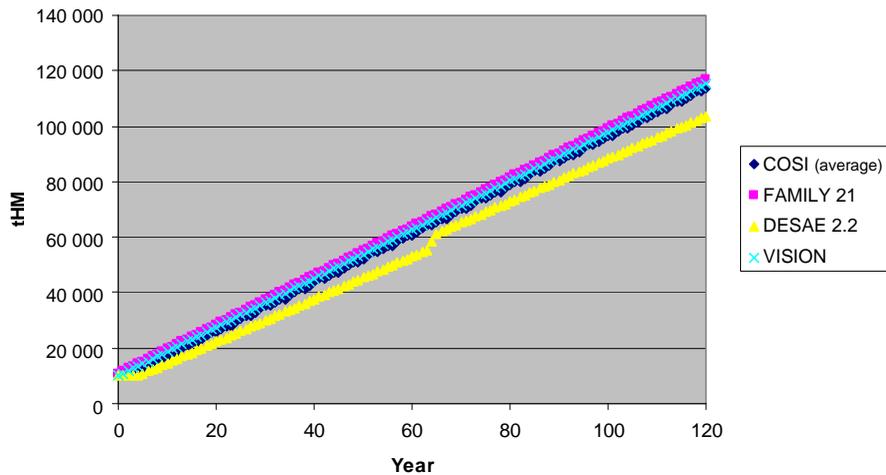
The results given by the codes are very close for mining, enrichment and fuel fabrication.

A small difference appears in the LWR UOX irradiated fuel inventory, due to an accumulation effect. For instance, at the end of Scenario 1, the results given by the codes are:

- COSI – 113 700 tonnes;
- FAMILY 21 – 116 905 tonnes;
- VISION – 115 326 tonnes;
- DESAE 2.2 – 103 627 tonnes.

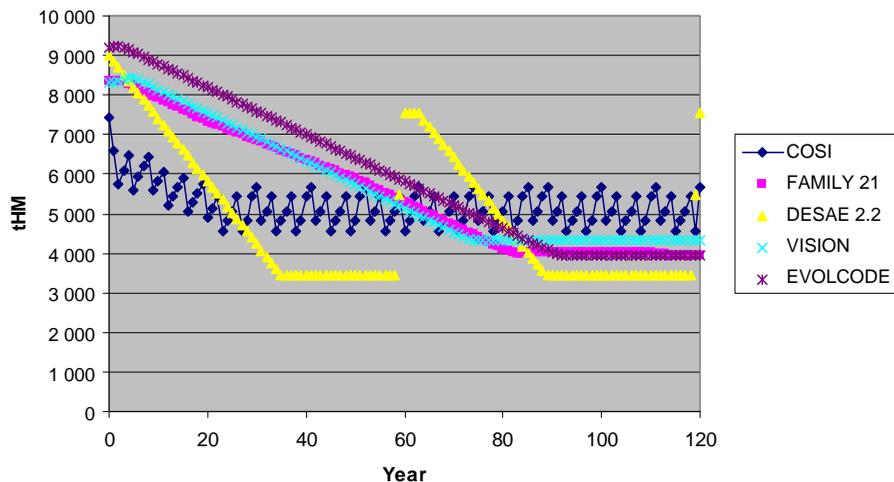
These differences come both from initialisation of spent fuel storage at the beginning of the calculation and from small differences on annual spent fuel unloading. For instance, in the FAMILY 21 calculation, the total LWR UOX irradiated fuel inventory at year 120 is higher because of annual unloading which is a little bit higher than COSI and VISION.

The year of the first unloading of spent fuel also has an impact on the accumulated LWR UOX irradiated fuel.

**Figure 4: Scenario 1 – LWR UOX irradiated fuel inventory**

### Scenario 2

Among all the contributing codes, COSI, FAMILY 21, EVOLCODE and VISION calculate the same values for natural uranium needs, SWU needs, UOX and MOX fuel needs. For DESAE 2.2, natural uranium needs, SWU needs, UOX and MOX fuel needs are lower. The reason could be a difference in core loading assumptions. The results on the back end of the fuel cycle are presented in Figure 5.

**Figure 5: Scenario 2 – LWR UOX irradiated fuel inventory**

For UOX spent fuel inventory, the results are very different among all the codes. Until approximately year 25, the decreasing slope of all results (except DESAE 2.2) agrees very well, according to the difference between the reprocessing requirement assumption (850 tHM of spent fuel per year) and the amount of spent fuel unloaded from LWR-UOX (790 t of spent fuel initial heavy metal per year). The different offsets of the curves come from differences in the first year of spent fuel unloading. For instance, between EVOLCODE and VISION, this difference is one year and leads to an offset of approximately 790 tHM. For COSI, the first year of unloading is year 3, with a difference of two years with VISION and three with EVOLCODE 2. In Scenario 3 the COSI calculation has been adapted (as COSI adjusted) to have the same initialisation as the VISION calculation. In that case, COSI, FAMILY, EVOLCODE and VISION are coherent.

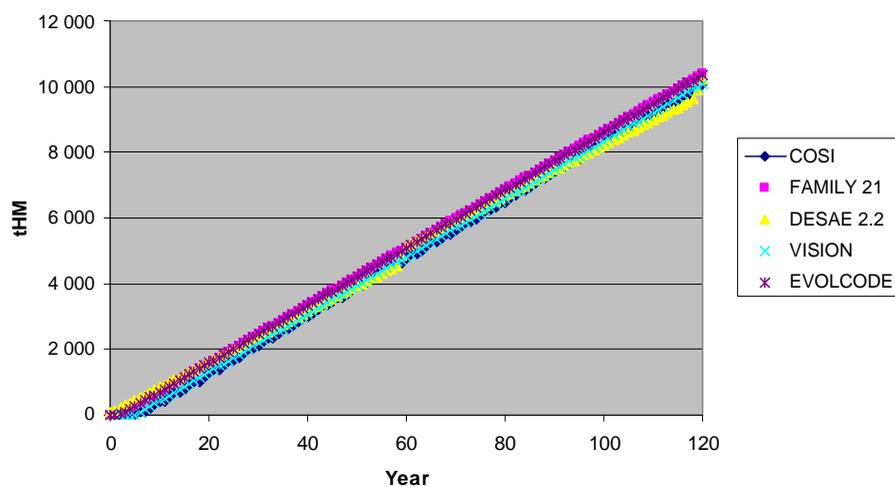
Beyond year 25, COSI reaches equilibrium in the spent fuel inventory due to a lack of spent fuel availability. For FAMILY, EVOLCODE and VISION, this stabilisation takes place at different years due to the differences mentioned above in the first unloading. The level of stabilisation is around 4 000 tHM, corresponding to the mass of spent fuel in cooling before reprocessing. For COSI, the averaged level is around 5 000 tHM and the minimum value is around 4 500 tHM. This difference is due to the fact that COSI takes into account the real cycle length of the reactors, 16.4 months. Thus, the mass of spent fuel unloaded is equal to 1 085 tHM. Moreover, the reprocessing is annual and occurs at the middle of the year. For these reasons, the spent fuel in cooling can be higher than 4 000 tHM.

For DESAE 2.2, the step at year 64 results from the unloading of the first cores of the reactors renewing the previous fleet of reactors. The renewing occurs at the same year for all the reactors of the park as in Scenario 1. However, the minimum spent fuel inventory is about 3 500 tonnes and should be at least equal to:

$$5 \text{ (years of cooling)} * 738.6 \text{ (annual unloading in DESAE)} = 3\,693 \text{ tonnes}$$

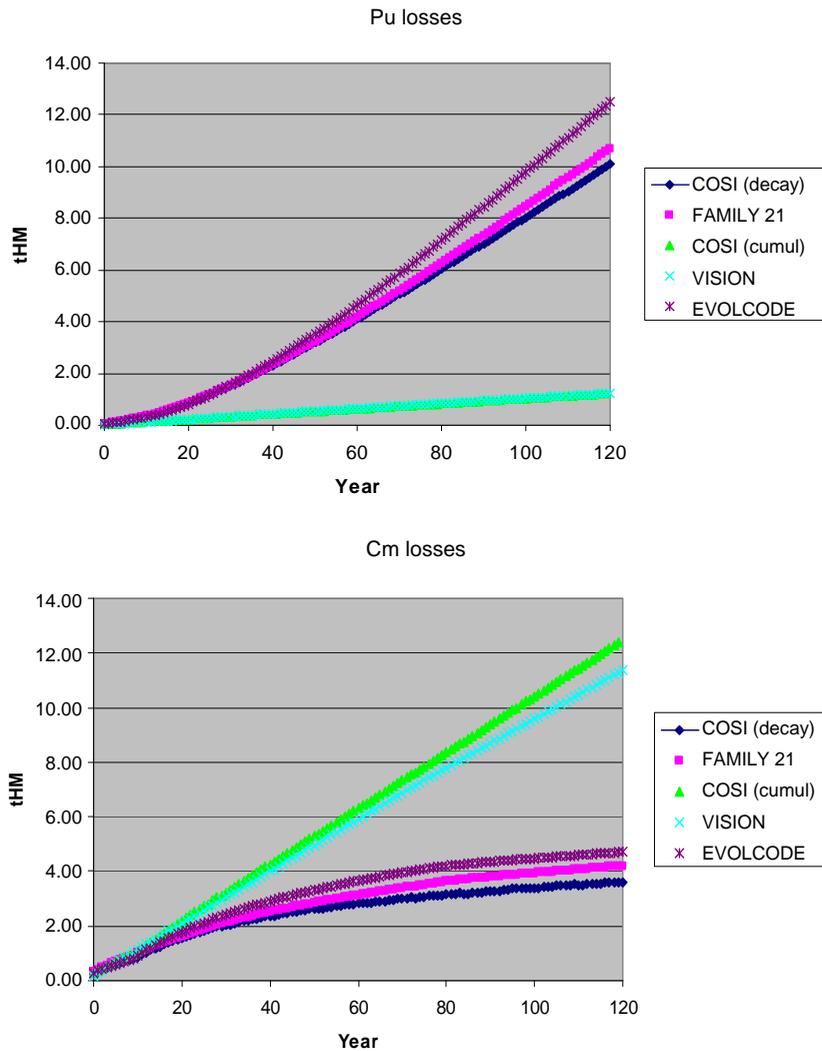
Moreover, some differences between codes are simply due to reporting results; some codes give the value at the end of a year and others at the beginning. Thus, a one-day difference (31 December versus 1 January) can appear as a full year difference.

**Figure 6: Scenario 2 – LWR MOX irradiated fuel inventory**



The differences in MOX spent fuel inventory are a consequence of differences in MOX annual fabrication since MOX fuel is not reprocessed in this scenario, and differences in the year of first MOX unloading: year 3 for EVOLCODE, year 5 for COSI, 6 for VISION, 2 for FAMILY 21 and year 0 for DESAE.

The amount of plutonium present in the waste is shown in Figure 7. Two groups of results can be seen in the figure. The first includes the simulations without decay of the waste: COSI (CUMUL) and VISION. The second group includes COSI (DECAY), EVOLCODE, FAMILY and takes into account the decay. For the latter group, the main contribution comes from the decay of the  $^{244}\text{Cm}$ , leading to  $^{240}\text{Pu}$ . The reasons for the larger amount of Pu in the EVOLCODE simulation are a larger creation of Cm during the irradiation and a 7% higher amount of annual spent fuel reprocessed.

**Figure 7: Scenario 2 – Pu and Cm losses**

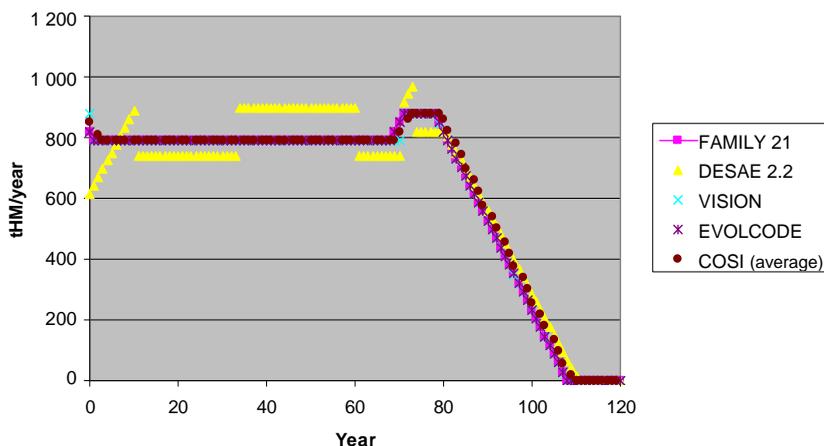
Thus, in order to explain the differences between codes for the amounts of Pu and minor actinides in the waste, it is necessary to consider four factors:

- 1) A different reprocessing amount of heavy metal: this amount is higher for DESAE 2.2, FAMILY 21, EVOLCODE and VISION than for COSI. With COSI, a lack of spent fuel for reprocessing appears by year 22.
- 2) In COSI and EVOLCODE, the reprocessing strategy of “oldest batch are reprocessed first” is applied. However, due to the lack of spent fuel availability detected in COSI, the effective decay before reprocessing of spent fuel is different.
- 3) Each code has applied its own neutron spectra and cross-sections, leading to slightly different isotopic compositions in the spent fuel.
- 4) COSI, EVOLCODE and FAMILY codes account for the decay of nuclear waste after reprocessing, whereas DESAE and VISION do not consider this decay.

### Scenario 3

The five codes contributing to Scenario 3 are COSI 6, FAMILY 21, DESAE 2.2, EVOLCODE and VISION. Figure 8 presents the LWR UOX fuel fabrication.

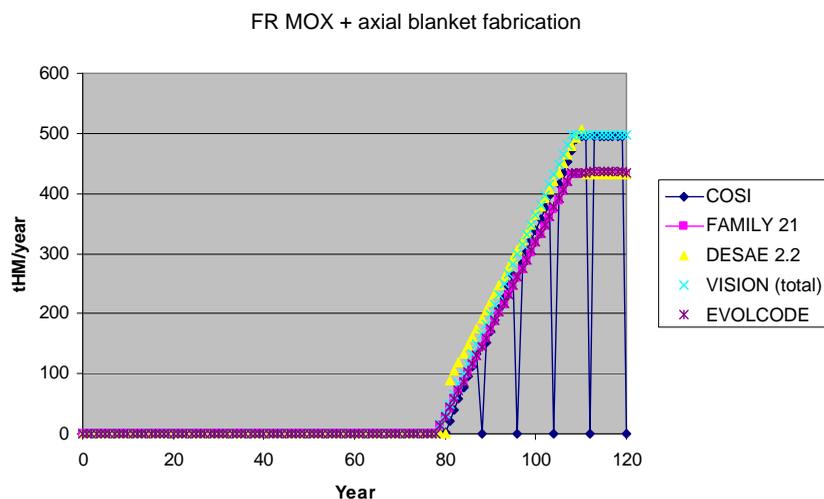
**Figure 8: Scenario 3 – LWR UOX fabrication**

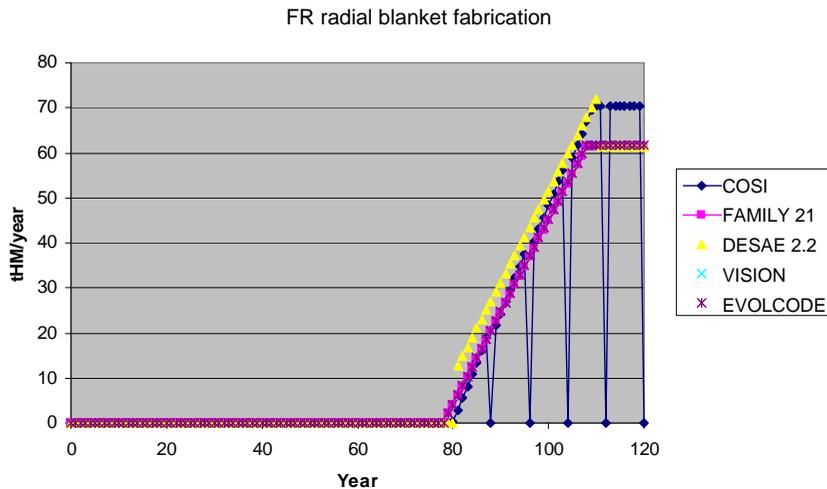


Among all the contributing codes, COSI, FAMILY 21, EVOLCODE and VISION calculate the same values for UOX fuel needs. For DESAE 2.2, LWR UOX needs are different. An important difference appears from year 34 to year 60 and remains unexplained.

Figure 9 presents the results on the fast reactor fuel and blanket fabrication. For fast reactor fuel and blanket fabrication, the apparent differences between codes can be explained. All the codes exhibit the same trend. However, COSI gives a value equal to 0 for the years 88, 96, 106, 112 and 120. In COSI, all the fast reactor fleet is simulated with one single fictive reactor with a variable power. Due to the cycle length for fuel and blankets which is different from one year, there is no reloading every eight years in the fast reactor. It is also important to note that VISION cannot separate fuel, axial blankets and radial blankets in the simulation and the results indicate the total of all the contributions.

**Figure 9: Scenario 3 – fast reactor fuel and blanket fabrication**



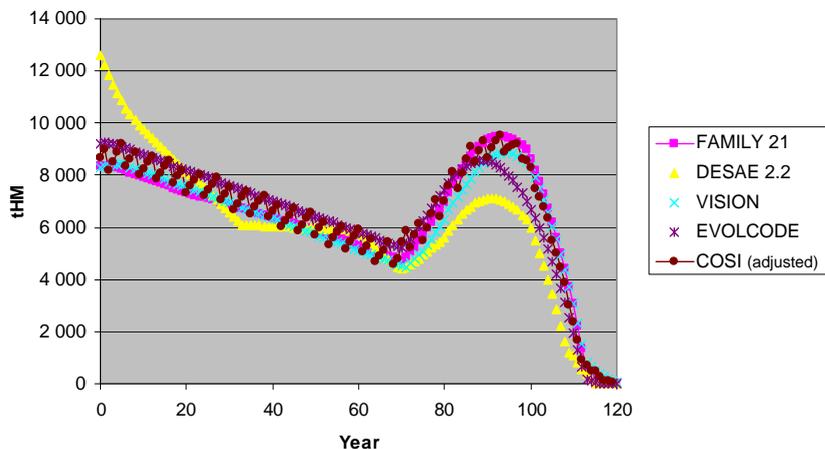
**Figure 9: Scenario 3 – fast reactor fuel and blanket fabrication (cont.)**

If we calculate the annual average of the values after the deployment of the reactors, we find a discrepancy of only 1% among all the codes for FR MOX + axial blanket, and again a 1% for the radial blanket. Table 3 summarises the average annual values at equilibrium period.

**Table 3: Scenario 3 – fast reactor annual average fabrication results**

	COSI	FAMILY 21	VISION 2.2	EVOLCODE	DESAE 2.2
Annual average MOX + axial blanket fabrication after 2100	435.0 tonnes	431.8 tonnes	–	435.4 tonnes	431.8 tonnes
Annual average radial blanket fabrication after 2100	61.8 tonnes	61.3 tonnes	–	61.8 tonnes	61.3 tonnes
Total	496.8 tonnes	493.1 tonnes	497.4 tonnes	497.2 tonnes	493.1 tonnes

As for LWR UOX irradiated fuel inventory, the results indicate a good agreement by taking into account the feedback from the Scenario 2 comparison and after several iterations for the tuning of the initial conditions (see Figure 10).

**Figure 10: Scenario 3 – LWR UOX irradiated fuel inventory**

## Conclusions

The results and the analysis of the calculations lead to the following conclusions:

- 1) The general trends observed for each code are the same for the three scenarios calculated in the benchmark.
- 2) All the scenario codes give very close results for the first scenario. However, this scenario is very simple and cannot serve as a reference for the comparison of the codes.
- 3) When the level of complexity of the scenario increases, the general trends observed for each code are the same but some discrepancies appear. The comparison of the results on the second scenario demonstrates the importance of initial assumptions and the common interpretation of the hypotheses and results in the comparisons.
- 4) A tuning of the assumptions is always necessary. This necessity is due to the difference of interpretation for initial conditions and to some missing assumptions which may appear. Thus, several iterations can be necessary to converge.
- 5) Once the tuning and iterations have been made, some remaining discrepancies subsist and result mainly from the capacity of modelling of the codes, the transition period in the third scenario, the differences in physical models (irradiation and depletion calculations) and the flexibility offered by the different codes.

However, some discrepancies will remain whatever the level of the analysis and the number of iterations, because of the different methods of calculations. This benchmark was limited to the comparison in heavy element material flows. A comparison for isotopes would have probably led to other discrepancies and would have necessitated a more detailed investigation on the physical models used by the codes.

## Reference

- [1] Nuclear Energy Agency (NEA), *Benchmark on Scenario Codes*, OECD/NEA, Paris, forthcoming.



## Industrial research for transmutation scenarios

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### Abstract

*This paper presents the results of research scenarios for americium transmutation in a 22<sup>nd</sup> century French nuclear fleet, using sodium fast breeder reactors. We benchmark the americium transmutation benefits and drawbacks with a reference case consisting of a hypothetical 60 GWe fleet of pure plutonium breeders. The fluxes in the various parts of the cycle (reactors, fabrication plants, reprocessing plants and underground disposals) are calculated using EDF's suite of codes, comparable in capabilities to those of other research facilities. We study underground thermal heat load reduction due to americium partitioning and repository area minimisation. We endeavour to estimate the increased technical complexity of surface facilities to handle the americium fluxes in special fuel fabrication plants, americium fast burners, special reprocessing shops, handling equipments and transport casks between those facilities.*

## Introduction

Transmutation studies have been performed since the early 1970s with the Superfact experiment in Phénix [1]. The 1991 and 2006 French waste laws have encouraged the academic and public research centre communities to pursue research in long-term nuclear fuel cycle scenarios, using transmutation. Industry also participates in the debate and follows the 2006 law by performing R&D with its own tools and means. Since it has industrial facilities to run on a day to day basis, it incorporates in the numerous scenario studies some of its actual industrial practices, keeping in mind that simplicity is often a key success factor in technology deployment. To that purpose, we present here the results of scenario studies for americium transmutation in a 22<sup>nd</sup> century French nuclear fleet, using sodium fast reactors (FR).

We describe briefly the current EDF fuel management system for its 58 LWR reactors fleet. We then give the main technical parameters for the americium scenarios studied as well as comparisons to the reference case consisting of 60 GWe of plutonium fast breeders. The simulation tools are then briefly described, and we subsequently move to the subject of results in terms of actinides (major and minor) in the whole cycle, as well as the consequences for underground disposal. Particular emphasis is placed on underground thermal load, underground disposal area and volume for the inventories produced around 2150. Finally, we endeavour to draw the technical and resource balance between underground actinide management and surface actinide management.

## The industrial fuel management system for the current LWR fleet

### *Nuclear production*

There are currently 58 PWR reactors operating in France, representing 63 GWe and designed on standardised PWR reactors series. EDF aims at increasing the lifetime of its Generation II reactors over 40 years. Further, EDF is currently building the EPR reactor, a 1 650 MW unit, on the Flamanville site.

### *The current fuel cycle: Spent fuel and back end*

The French fuel cycle has already been presented in detail in Ref. [2]. In the 1970s, in order to preserve varied options for the future, including the development of FR, the decision was taken to develop a closed fuel cycle based on reprocessing of spent fuel and retrieving plutonium and uranium at the La Hague plant operated by AREVA NC. High-level waste (fission products and minor actinides – HLW) is confined into glass canisters and reusable nuclear materials (plutonium and uranium) constitute the potential energy resource to be recycled. Nuclear spent fuel reprocessing and recycling in France has reached the stage of mature industrial practice and represents a first step towards the sustainability of nuclear power while assuring a secure treatment and conditioning of HLW and preserving resources of fissile and fertile fuels. The main figures of the current fuel cycle at equilibrium are presented in Ref. [2]. From 2010 on, EDF envisages increasing from 850 t to more than 1 000 t the quantity of spent fuel reprocessed every year so that the annual amount of MOX fuel will increase up to 120 t. Today, 22 900 MW-class PWR are licensed for MOX fuel and a licensing procedure is under way for two extra reactors. The spent MOX will be reprocessed in the future: it constitutes a resource of nuclear fuel for the long term that can be reused especially in future FR to meet future energy needs. This strategy allows for time, and can be extended over the coming decades, while preparing for future options.

### *The French Act of 28 June 2006 on “A Sustainable Management of Nuclear Materials and Radioactive Waste”*

As for the management of long-lived and high-activity nuclear waste, after 15 years of research and studies of different options, the Act of 28 June 2006 established a national strategy for managing nuclear materials and radioactive waste. A stepwise programme for long-lived waste

management (high- and medium-activity) is defined along various approaches: a retrievable geological repository as a reference solution (authorisation decree in 2015, beginning of operation in 2025); the creation of new facilities for interim storage in 2015, the pursuit of research on partitioning and transmutation (P&T). More precisely, for P&T, the act requests the assessment of industrial prospects of Generation IV systems and accelerator-driven systems (ADS) for presentation to the French Parliament in 2012. The assessment should account for the potential benefits of P&T (i.e. minor actinide – MA – recycling or not) for the long-term management of HLW and also for their impact on future fuel cycle facilities and reactor operation in terms of safety, radiation protection and costs. The act foresees the commissioning of a prototype (reactor and cycle) in 2020, which triggers R&D on a new generation of FR and advanced recycling modes (with or without MA). It further states that, whichever chosen strategy for the management of waste, industrial interim storage of spent fuel and deep geological disposal remain necessary.

### ***To transmute all the minor actinides or americium only?***

Scenario studies compare many transmutation options. In particular, one has the choice to transmute all the MA – Am, Cm, and Np – or Am only.

Np transmutation is not of any real interest. This actinide is not mobile under the chemical conditions in the French geological site for the repository, and thus it has no environmental impact. Its decay heat is low and there is no real benefit to the underground repository area.

As for Cm transmutation, due to the short Cm half-life (18 years), it has a weak impact on an underground repository area. The only significant gain would be with regard to the waste radiotoxicity in the repository but on one hand, this gain is variable a long time and it is not different from the gain obtained with Am alone transmutation after 100 000 years. On the other hand, radiotoxicity is a questionable indicator as we will see later on in this paper. As for drawbacks, Cm handling in the fuel cycle is very hazardous; in particular, the thermal load and the level of neutron sources at the fuel fabrication stage are very high as compared to the FR MOX standard fuel fabrication. Besides, the decay heat and neutronic sources of spent MA-bearing fuel are very high: this has a huge impact on fuel handling and the power plant availability as well as the transportation of spent fuel. That is why EDF is in favour of considering in research scenarios transmutation of Am only and to dispose of Cm in the geological repository after an interim storage for decay heat. Thus, in this paper, only Am transmutation research scenarios are considered although we also perform the studies of scenarios where all MA are transmuted for comparison and checking purposes.

### **Characteristics of the various transmutation scenarios**

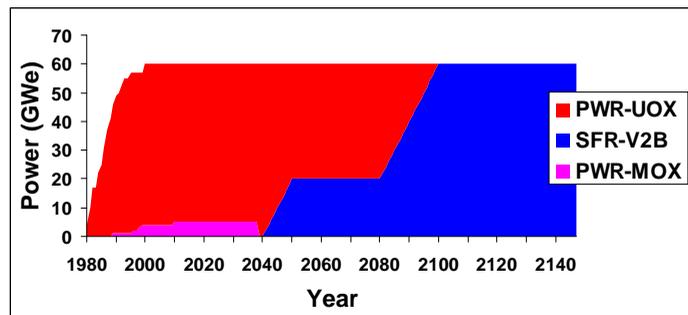
The principle of the study is to define a reference scenario of FR deployment without MA transmutation, and to compare it to other scenarios built on the same framework, but with various Am transmutation options.

#### ***The plutonium reference case***

The reference case (scenario S\_ref) is based on the EDF prospective view of the transition from its current nuclear fleet and a Generation IV FR fleet throughout the 21<sup>st</sup> century, as shown in Figure 1.

The current reactors' renewal starts as from 2020, with Generation III PWR (EPR type). Generation IV FR deployment is supposed to start in 2040, and the Pu monorecycling in PWR to stop a few years earlier. With a kinetics of 2 GWe replaced each year, and an installed nuclear power fixed at the current level, 60 GWe, 20 GWe are replaced in 2050. The fleet is then composed of two-thirds of Generation III PWR, and one-third of Generation IV FR. Generation III PWR are then replaced by FR as from 2080, and the fleet is thus entirely composed of FR in 2100. All MA are vitrified with the fission products.

Figure 1: SFR deployment scenario



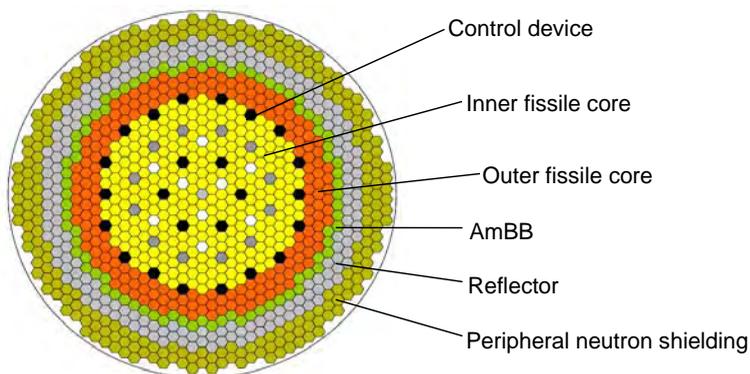
The Generation IV FR concept used has been developed by the CEA and is known as SFR V2B [3]. Its core (Figure 2) contains 453 fuel assemblies (FA), with ~16% mean Pu content, for a 3 600 MW thermal power. The core Pu inventory is ~74 tonnes. This design is characterised by a breeding gain a little more than zero, allowing isogeneration, without any fertile blanket. The core residence time of the fuel is 7 years (5 cycles lasting 410 equivalent full power days with a 81% loading factor). The ex-core time is also 7 years (assumption we also used for PWR fuel): 5 years of minimum cooling time before used fuel reprocessing, and 2 years after, before in-core loading. Thus the deployment of SFR V2B requires about 17 tonnes of Pu per GWe. The Pu necessary to feed the 20 GWe of SFR deployed between 2040 and 2050 is first extracted from UOX and PWR MOX used fuels. SFR MOX fuel reprocessing is supposed to start only when the stock of PWR MOX used fuels is completely exhausted (i.e. in 2059).

### The Am transmutation scenarios

The Am transmutation scenarios are based on the same framework. Two main options are considered, the homogeneous transmutation (scenario S\_hom), whereby Am is mixed with Pu in the standard SFR fuel, and the heterogeneous transmutation, whereby Am is mixed with depleted uranium in radial blankets disposed outside the fissile core [4].

The main advantage of the heterogeneous option is to reduce the number of Am fuel bearing assemblies with respect to the homogeneous one, more currently studied. Figure 2 shows the position of 84 Am-bearing blanket assemblies.

Figure 2: SFR V2B core with AmBB (heterogeneous scenario)



A homogeneous transmutation core would necessitate 453 Am-bearing FA. Taking into account the different loading schemes of homogeneous FA and heterogeneous AmBB assemblies, it is possible to show that the homogeneous scenario requires an Am-bearing FA flux 5 to 10 times higher than the heterogeneous scenario (1 300 Am-bearing core assemblies per year versus 120 to 300 AmBB assemblies per year in the case of 20 SFR V2B reactors loaded).

Those Am-bearing assemblies will have to be fabricated in remote control plants, requiring maintenance and repair with telemanipulators or robots. Reducing their output and size will therefore be a big advantage, if only from a technical point of view to begin with. It can also be argued that the safety demonstration, while not impossible in homogeneous cores, will be more difficult not so much because of neutronics parameters [5] but because of gas production in the actinide-bearing FA. The drawback of this choice is to raise the Am content of Am-bearing assemblies. A 20% content is required to limit the required number of AmBB while efficiently transmuting the Am. AmBB irradiation time is 14 years, to ensure a convenient transmutation rate (35%), and the AmBB assemblies have to be rotated 180° after half this time.

In the case of the homogeneous option, all the FR are supposed to be loaded with standard assemblies in order to keep the Am content as low as possible. In the case of the heterogeneous option, the fraction of FR equipped with AmBB is not fixed, and depends on the amount of Am to transmute and the numbers of FR available. Two options can then be considered:

- All Am contained in the used assemblies reprocessed as from 2038, when FR fuel fabrication starts, have to be transmuted in AmBB, meaning that all the FR have to be loaded with AmBB if necessary (scenario S\_het); in this case, reduction of repository surface area due to lower heat content is considered more important than the difficulties of loading all the FR with AmBB and running the fabrication plants at high flux.
- Only a fraction of the SFR, for instance 50%, is loaded with AmBB (scenario S\_het\_50%). In this case, the penalty of loading all the FR with AmBB and running larger fabrication plants at high flux is considered greater than the benefits of repository surface reduction due to lower underground heat content. The number of FR used for transmutation is reduced by half, and the Am which cannot be loaded in the reactors when partitioning starts in 2038 is sent directly to the waste stream.

The SFR adapted to the Am transmutation are called FR 1500<sub>Am</sub>. The other FR 1500<sub>Pu</sub> are pure Pu breeders and do not have the special equipments necessary for AmBB loading. This option projects in the future the current LWR EDF fleet where only 24 reactors out of 58 have the MOX loading capability from the technical and licensing point of view. For first order calculations a 60 GWe fleet around 2100 is composed of 40 FR 1500. Those data are valid for research scenarios. In industrial practice, EDF does not consider Am partitioning from PWR MOX fuels in 2038. MOX fuels can be industrially reprocessed with an adapted PUREX process.

The last scenario we considered is based on the scenario S\_het, but with a different assumption about the date of implementation of the Am transmutation, putting it off from 2040 to 2080, date of the second phase of SFR massive deployment. The first motivation for this assumption is to propose a more progressive scenario. Indeed, SFR deployment at an industrial scale is really a big challenge. The Am option, requiring the implementation of advanced reprocessing and the fabrication and handling of a more complex fuel, is another big technical challenge; it thus seems more realistic to postpone its implementation to a later date, when the SFR fleet is strongly installed and the closure of the fuel cycle with SFR used fuel reprocessing is well established.

In addition, by doing so, we limit the Am transmutation to Am issued from SFR used fuel: in that case, Generation IV FR would minimise their own HLW production, and we consider that the MA produced before, especially by PWR MOX fuel, will be sent to the repository. Besides, restraining the Am transmutation to the Am produced by the SFR fleet makes possible to limit the proportion of SFR loaded with AmBB, almost at 50% like in the S\_het\_50% scenario, as we will see.

## Simulation tools and methodologies

### Simulation tools

Two tools are used for the complete simulation of a scenario. First, the scenario simulation code TIRELIRE-STRATEGIE [6] simulates the operation of the nuclear fleet and the associated fuel cycle

facilities. Then, exploiting the results of this simulation (i.e. for each year, uranium consumption, the mass flows, fuel inventories and isotopic compositions in all fuel cycle facilities, and the number of HLW packages together with their thermal power outputs, etc.), our final HLW repository model calculates the minimal repository area requested for disposing of the waste packages. The vitrified waste package repository design issues from the ANDRA 2005 report [7]. The area minimisation process is driven by the temperature constraint fixed by ANDRA; the temperature in the clay layer must be kept below 90°C. Repository thermal calculations used for this optimisation are performed with the SYRTHES code, developed by the EDF R&D division [8].

### **Detailed criteria for comparisons of scenarios**

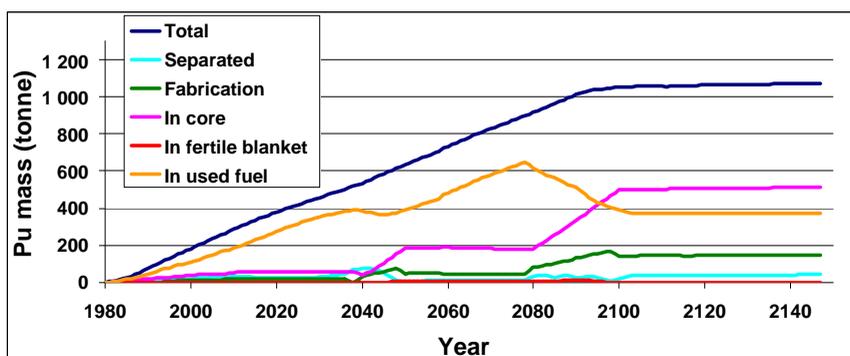
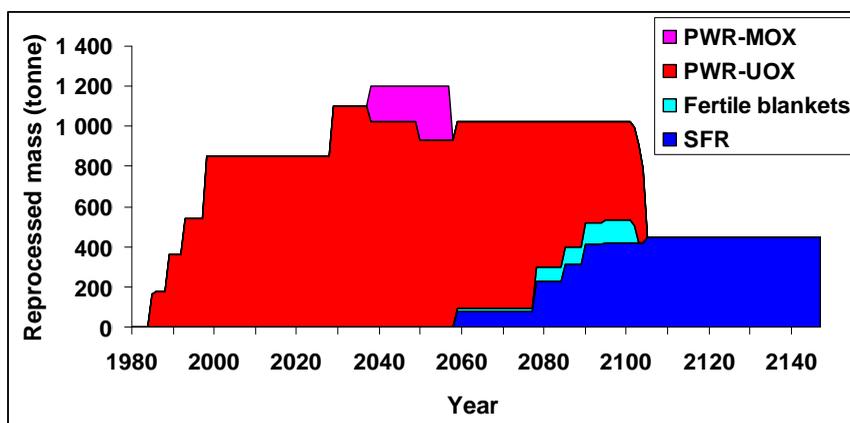
MA transmutation is a complex process and the number of criteria to assess can be very large, which can finally result in an inefficient assessment. Criteria selection is therefore an important step of scenario studies. The main criteria chosen are the following ones: inventories and characteristics of materials and waste induced in the different scenarios (in the fuel cycle including reactors and in the geological repository); facilities' characteristics and transportation needs; impacts on geological repository (underground area, safety); impact on radiation protection of citizens and workers; economical performance (cost of each scenario); impact on power plant (safety, availability, cost); industrial risk; fuel resource consumption. One of the criteria very often emphasised to justify transmutation is the potential radiotoxicity of waste. It is actually a criterion very difficult to deal with and not so pertinent. The radiotoxic inventory of a waste quantity corresponds to the dose received by a set of people who would have ingested this waste quantity. It is a theoretical indicator which has a real meaning only if there is a path from the waste to the set of people. In the case of the geological repository, for the site considered in France, MA are not mobile and so do not move out of the repository. In fact, ANDRA, in charge of the repository studies, has performed a safety assessment which shows that the environmental impact is due to the fission and activation products and is very low, between one and two orders of magnitude under the regulatory limit. Indeed the dose at the outlet stays below one microsievert per year, even at its maximum reached after ~500 000 years in the normal evolution scenario and below one microsievert per year, maximum reached after 25 000 years, in the altered evolution "borehole" scenario. The main contributors for vitrified waste are  $^{129}\text{I}$  and  $^{36}\text{Cl}$ . Pu isotopes play a role only in case of the altered evolution "borehole" scenario, but their contributions are at least three orders of magnitude below (maximum reached after 100 000 years). No MA contributions are quoted at the outlet [7]. Therefore EDF does not consider potential radiotoxicity as a pertinent indicator to justify MA transmutation unless the French safety regulator, after the advice of its standing advisory group ("Groupe Permanent Déchets"), requires it for technical reasons not foreseen at this stage.

### **Detailed results and consequences on fuel disposal**

#### **Reference scenario**

The cumulated amount of Pu requested for feeding a fleet of 60 GWe of SFR V2B is about 1 070 tonnes (Figure 3). Its repartition is 500 tonnes in core, 150 tonnes in fabrication facility, 370 tonnes in used assemblies waiting for cooling, and a few tens of tonnes of Pu separated and stored as a reserve for fabrication. To get such a Pu mass in 2100, it is necessary to make use of fertile blankets during all the transition period (they are not useful later, the last one being reprocessed in 2101). Lower axial blankets are enough, but they must be loaded in all the SFR operated during the transition period. On the whole, 2 700 tonnes of fertile blankets are needed, producing about 130 tonnes of Pu.

In order to get the 350 tonnes of Pu necessary to deploy the first 20 GWe of SFR, it is necessary to increase the amount of reprocessed used fuel to a maximum value of 1 200 tonnes per year, of which 270 tonnes originate from PWR MOX fuel in 2050 (Figure 4).

**Figure 3: Reference scenario, Pu mass inventories in the fuel cycle****Figure 4: Reference scenario, reprocessed fuel masses (TMLi)**

The ratio Pu/U+Pu in that case is slightly higher than the 20% limit currently admitted in the La Hague facility. The reprocessing of the SFR MOX fuel starts in 2059. Therefore, it would be possible to use the same reprocessing technology till this date, facilitating the transition towards the SFR fuel cycle. After 2055, all the PWR used fuel is exhausted, and the annual mass of SFR MOX fuel to be reprocessed each year is stabilised at about 450 tonnes per year.

## **Am transmutation scenarios**

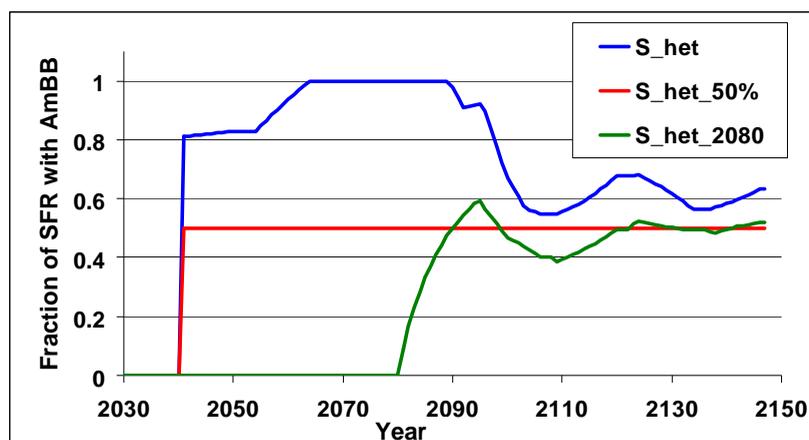
### *Plutonium and minor actinides inventories*

The Pu content has been adjusted in the case of the reference scenario in order to ensure Pu isogeneration. Am transmutation increases the breeding gain of the fissile core in case of homogeneous transmutation, or by Pu production in the AmBB in case of heterogeneous transmutation, but it is not enough to do without the fertile blankets. In order to simplify the simulations, we kept for all the scenarios the same assumption about using 2 700 tonnes of fertile blankets during the transition period. Table 1 summarises the results obtained at the end of the scenario (2150). Due to their higher breeding gain, the transmutation scenarios feature higher Pu cycle inventory at equilibrium than the reference one, the extra Pu being stored. At equilibrium, the Am content in the case of homogeneous transmutation is 0.8%, but there is a peak at ~3% at the beginning of SFR deployment in 2040, due to the important amount of Am accumulated in the PWR used fuel (especially the MOX ones).

**Table 1: Plutonium and minor actinides inventories in 2150**

Scenario		S_ref	S_het	S_het_50%	S_het_2080	S_hom
Am content	Equilibrium	–	CCAm 20% Am	CCAm 20% Am	CCAm 20% Am	0.8% Am
	Maximum					2.8% Am in 2040
Pu (tonnes)	Cycle	1 070	1 240	1 200	1 150	1 120
	Waste	20	25	25	23	30
MA (tonnes)	Cycle	18	105	95	95	55
	Waste	390	155	210	232	155

Considering the heterogeneous transmutation scenarios, S\_het assumes the transmutation of all the Am issued from the used fuel reprocessing, and thus requires loading AmBB in all the SFR available between 2064 and 2090. After this date, the proportion of reactors equipped with AmBB decreases progressively (with some oscillations) towards the equilibrium value, 55%, which is not reached before 2200 (Figure 5).

**Figure 5: Proportion of SFR V2B equipped with AmBB**

In the case of the other heterogeneous options, S\_het\_50% imposes a 50% proportion, while S\_het\_2080 starts the Am transmutation in a situation already close to the equilibrium, thus limiting the proportion around 50%. Of course, this means that a significant amount of Am has been vitrified during the transition period: 55 tonnes in case of S\_het\_50%, 80 tonnes in case of S\_het\_2080. On the other hand, almost all the Am issued from the SFR used fuel reprocessing is transmuted.

Am transmutation results in a significant increase of the MA inventory (mainly Am) in the fuel cycle: 55 tonnes in case of homogeneous transmutation, around 100 tonnes in case of heterogeneous transmutation, compared to less than 20 tonnes in case of the reference scenario.

Am transmutation allows a significant reduction of the amount of MA sent to the final disposal. The repository transuranium element (TRU) inventory is about 400 tonnes in 2150 in case of the reference scenario, ~180 tonnes in case of the S\_het and S\_hom scenarios (reduction factor higher than 2). The performance of S\_het\_50% and S\_het\_2080 scenarios is lower, with a reduction factor above 1.5.

On the other hand, if we consider the TRU total amount, both in cycle and repository, it is important to note that the reduction factor is very limited, due to the huge amount of Pu in the fuel cycle.

**Table 2: TRU inventories (waste and total)**

Scenario		S_ref	S_het	S_het_50%	S_het_2080	S_hom
TRU (t)	Waste	410	180	235	255	185
Reduction	Waste	1	2.3	1.7	1.6	2.2
TRU (t)	Total	1 500	1 355*	1 400*	1 440*	1 310*
Reduction	Total	1	1.11	1.07	1.04	1.15

\* For the transmutation scenarios, we did not take into account the extra Pu due to the over-breeding.

### Impact on waste disposal

The total number of vitrified waste packages produced in the various scenarios is almost the same, a little more than 140 000. Am transmutation brings no benefit with respect to this parameter. It is due to the fact that the glass content is imposed by criteria related to fission products, not actinides (we have assumed that the current criterion limiting at  $10^{19}$  the maximum  $\alpha$  decay per gram of glass, integrated over the first 10 000 years of cooling, will be released beyond  $2 \cdot 10^{19}$   $\alpha/g$  according to CEA R&D work [9], and thus will have only a marginal impact on the glass fabrication). Thus the number of glass canisters is practically proportional to the energy produced, which is the same for all the scenarios, with a rate of around two glass canisters per TWhe.

**Table 3: Transmutation impact on total vitrified waste package number, and on total repository area**

Scenario	S_ref	S_het	S_het_50%	S_het_2080	S_hom
Vitrified HL package (total number)	141 000	141 000	141 000	142 000	140 000
Vitrified HL package disposal area (normalise area $S_{ref} = 1$ )	1	0.55	0.66	0.71	0.57
Total repository area (HLW + MLW + underground common installations)	1	0.69	0.76	0.80	0.7

If the number of waste packages is the same, their heat power is quite different, according to their Am content. Considering a waste canister's interim storage lasting 60 years before their disposal, the underground area needed by a waste package issued from SFR MOX reprocessing is decreased by ~50% if the Am is transmuted. The reduction of the total repository area, including access drifts, compacted waste packages issued from the fuel structures compaction, or glass canisters issued from used fuel reprocessed before 2040, is smaller.

For research purposes only, we considered a unique repository for the whole duration of the scenarios. In case of the reference scenario with no Am transmutation, its global area should be around 30 km<sup>2</sup>. Thanks to Am transmutation (scenarios S\_hom and S\_het), the area needed for the glass canister would be reduced by ~45%, and the total repository area by 30%. If only half of the SFR are loaded with AmBB (scenario S\_het\_50%), these area are reduced respectively by 33% and ~25% and by 30% and 20% in case of the scenario postponing the transmutation to 2080. It must also be noticed that the repository area reduction will be most effective at the beginning of the 22<sup>nd</sup> century, the reduction factors discussed above being obtained in the 2150 time frame.

### Technical balance between underground actinide management and surface actinide management – A methodological approach

We first present here partial technical elements for the handling interfaces of AmBB assemblies between fabrication plants, the inside of the reactor vessel, the other power plant systems and the reprocessing plant. Am in fresh fuel results in increasing its power, neutron and gamma activity, hindering and complicating its fabrication and handling.

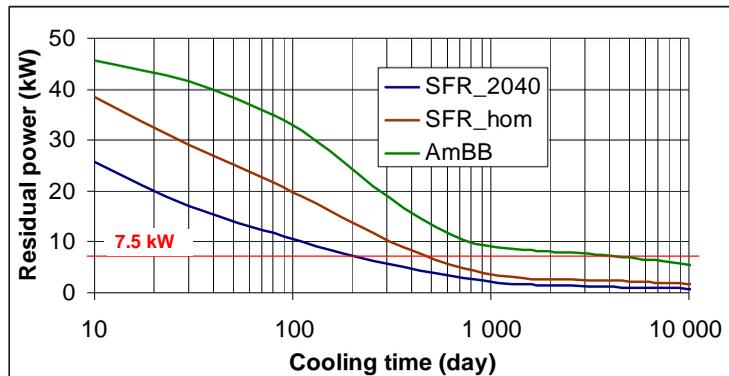
These consequences are enhanced in the case of AmBB, with 20% of Am. Am homogeneous transmutation is the option presenting the more limited impact, as illustrated by Table 4; with ~3% of Am when the transmutation starts in 2040, the assembly power is increased by 60%, and

**Table 4: Power and dose (contact and 1 m) for the fresh SFR assemblies and AmBB in 2040**

	SFR (2040)	SFR_hom (2040)	AmBB
Power (kW/assembly)	0.7	1.13	2.7
Contact dose (mSv/h)	14.8	19.3	70
Dose at 1 m (mSv/h)	0.5	0.80	2.9

the total dose (neutron + gamma) at the bare assembly contact by 30% (but 60% at 1 m) with respect to the SFR V2B standard fuel. Nevertheless, the shielding would have to be reinforced both for fabrication and handling, including the reception and temporary storage at the power plant, and the need to use shielded cells for the MOX fabrication will have to be carefully investigated. On the other hand, shielded cells and remote control are very likely to be necessary for the AmBB fabrication, with a dose 5 to 6 times that of the FR MOX assembly, also with an increase of the fuel power density by a factor 6 with respect to the standard fuel. AmBB handling, transport and reception would require strong shielding measures.

Figure 6 shows the evolution with respect to the cooling time of the power of the irradiated assemblies. Maximum power currently admitted for discharging assemblies of the core is 20 kW, but R&D work in progress allows expecting this value to be increased to 40 kW. However, AmBB would still require 40 days of cooling before being unloaded. Before shifting the used assemblies to the reprocessing factory in a shielded cask, it is necessary to wash them, which requires a power less than 7.5 kW; it takes almost 10 years of cooling for AmBB residual power to decrease below this limit, and this should be done in an external storage tank filled with sodium so that operation of the reactor will not be hampered. This is so far the major costly step in the reactor facility for heterogeneous Am transmutation. In a previous paper [10], we proposed that an external sodium storage tank should be used for increasing the lower structure's inspection capabilities as an industrial prototype. It is expected that design modifications and successful R&D on visualisation techniques under sodium will allow for commercial FBR 1500<sub>Pl</sub> inspection of the lower mechanical structures without the need for core unloading in an external sodium storage tank.

**Figure 6: Assemblies' residual power evolution versus cooling time**

The feasibility of used AmBB transport over a long distance is not proven. Designing a cask even for transporting only one AmBB each time is a real technical challenge, taking into account its high level of residual power, power density, and also of neutron emission (~20 times the one of standard SFR V2B assemblies) which would impose reinforcing radiological protection with the consequence of increasing the difficulty to efficiently evacuate the heat and to limit the fuel clad temperature at a level ensuring their integrity. The reference solution in our heterogeneous transmutation studies is to let the AmBB assemblies cool down for 10 to 40 days inside the reactor vessel, store for ~10 years in an external sodium storage tank, wash and then cool down in a water pool at the reactor site until transport to the reprocessing site is feasible. For a site with two reactors and common sodium storage, this concerns less than 200 AmBB assemblies in a time frame of 14 years.

Though the homogeneous transmutation case looks easier, the residual power is doubled, and the neutron emission multiplied by a factor of 4 with respect to a standard SFR V2B assembly. Moreover, the number of used Am transmuted assemblies to be handled and transported each year is more than an order of magnitude higher than in case of heterogeneous transmutation.

It is therefore evident that further R&D work is necessary to reduce the scope of the technical options even for the single  $S_{het\_50\%}$  scenario chosen to illustrate the methodology of technical and economical balance between underground repository cost reduction and above the ground extra investment. Questions raised during this technical comparison will have to be answered in the scientific and technological phases of the R&D programmes. A classical example concerns the scientific milestones of the R&D programmes for partitioning and fabrication:

- partitioning of Am from a high-activity solution with 100 to 200 g of Am in the back extraction solution, all other actinides being sent to the HLW stream;
- demonstration of fabrication capability of 10 FR pins containing each 100-200 g of Am.

The corresponding fabrication milestone in the technological phase is the extrapolation to several (10 to 30) fuel assemblies containing between 250 and 300 such fuel pins.

As for the repository area, Table 3 shows that a gain of 24% with respect to the  $S_{ref}$  scenario could be reached in the  $S_{het\_50\%}$  scenario with only half of the FR loaded with Am, and transmutation starting in 2038. It is then theoretically possible to calculate a reduction in investment costs of the underground repository facilities induced by transmutation. On the other hand, transmutation will require investing in above-ground facilities. Economic evaluation is not possible at the present stage of transmutation research. On the other hand, and provided that the scientific feasibility of the major fuel cycle processes is achieved, it is possible to draw process flow sheets and define complexity factors for the process functions and equipments with respect to current industrial facilities of similar type. Both process flow sheets and complexity factors have to be validated in the technological feasibility phase. Taking the  $S_{ref}$  scenario as a reference case, one should define complexity factors for the supplementary facilities above the ground:

- The 20 FR  $1500_{Am}$  altered with respect to 20 FR  $1500_{Pu}$  in the reference  $S_{ref}$  scenario.
- The fabrication plant for AmBB assemblies, the reference being the FR MOX and fertile blanket plant for 40 FR  $1500_{Pu}$ , corrected for minor output flux differences when only 20 FR  $1500_{Pu}$  are used in the  $S_{het\_50\%}$  scenario.
- The modified reprocessing plants for Am partitioning and AmBB assemblies' reprocessing. The reference facilities include the MOX FR reprocessing plant, operating around 2060.
- The interfaces between the three main functions: reactors, reprocessing, fuel fabrication in the case of AmBB assemblies.

One would wish to know rapidly rough investment estimates. Even more important than those estimates which will not be known for at least 15 more years are the R&D resources which will be needed to consolidate the complexity factors and in particular investment in new R&D facilities. For the heterogeneous Am transmutation scenarios with a limited annual flux of AmBB assemblies at the industrial scale, R&D resources should be invested first in determining through reactor experiments and PIE the maximum feasible content of Am in the AmBB assemblies. Next, R&D resources should be invested in overcoming the 7.5 kW limit of FA residual power before sodium washing and designing long distance transport casks at high thermal load.

## Conclusions

For a utility like EDF, the most important objective in the 2006 waste law framework is to start bringing down from 2025 onwards its glass logs produced between 1980 and 2040 with the Marcoule and La Hague vitrification processes. They amount to several tens of thousands of glass logs with several tens of tonnes of minor actinides. Thus reducing the waste potential

radiotoxicity underground is not a repository criterion for EDF unless the French safety regulator, after the advice of its standing advisory group, requires it.

Several (industrial) research scenarios are studied for partial or total transmutation of Am in order to evaluate the benefits, mainly the heat load reduction in the repository and the drawbacks. Heterogeneous transmutation is preferred in research to reduce the size and complexity of AmBB assemblies' fabrication plants. Reduction factors of 5 to 10 for the annual fabrication fluxes with respect to the homogeneous transmutation case are obtained. The penalty is the high thermal power of these fuel assemblies and the more difficult handling techniques at all interfaces, in particular the fuel handling system above the reactor vessel. The other power plant systems, the external facilities and the transport also have to be considered in the analysis. Projecting 2009 industrial practices with MOX fuel in PWR, only half of an hypothetical 22<sup>nd</sup> century fleet of FR is equipped with Am transmutation capabilities and operated in that regime. Sensibility studies are performed by shifting beyond 2080 the beginning of the Am transmutation regime.

Further R&D work is needed to consolidate the numerous technical data, to progress in the evaluation of process flow sheets for the surface facilities and to raise the power limits at critical interfaces.

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## Requirements-driven comprehensive approach to fuel cycle back-end optimisation

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### Abstract

*It is desirable to devise a solution for the fuel cycle back end that is acceptable from the standpoint of public acceptance, as well as technologically and economically viable. While satisfactory technological solutions exist, they only address portions of the overall problem. A fully integrated effective solution satisfying all public concerns has yet to be developed. In particular, we aim to establish a comprehensive requirements-driven approach. To this end, requirements are defined for the high-level wastes with the intent not only to satisfy all technical constraints but also to make them “acceptable” to the public perception. Only then, the best mix of nuclear reactors, reprocessing and fuel forms is examined to determine an effective, viable overall system. One intended benefit is that there is no a priori bias for or against any specific nuclear system. In fact, a mix of several different systems will likely provide an optimum solution, promoting collaboration between the relevant industry and research entities in the fuel cycle back-end activities.*

## Introduction

When devising a long-term solution for the fuel cycle back-end, it is necessary to consider not only technical requirements and economics, but also acceptability from the societal viewpoint, including public acceptance. Technological solutions have been sought for and in many cases successfully developed to address specific issues and portions of the fuel cycle. However, a fully integrated solution satisfying all public concerns has yet to be developed.

Two major global requirements are: i) sustainability; ii) effective long-term management of spent nuclear fuel/nuclear waste. This paper focuses on the latter, which has also been the main focus of public attention and a *conditio sine qua non* for public acceptance. Traditionally, a search for an effective solution has focused on a specific reactor type and was driven by the fuel cycle front-end and core residence time. In this work we aim to establish a comprehensive approach where the search for a solution is driven from the fuel cycle back-end, with the primary intent of devising a system that generates “acceptable” wastes.

## US historical perspective

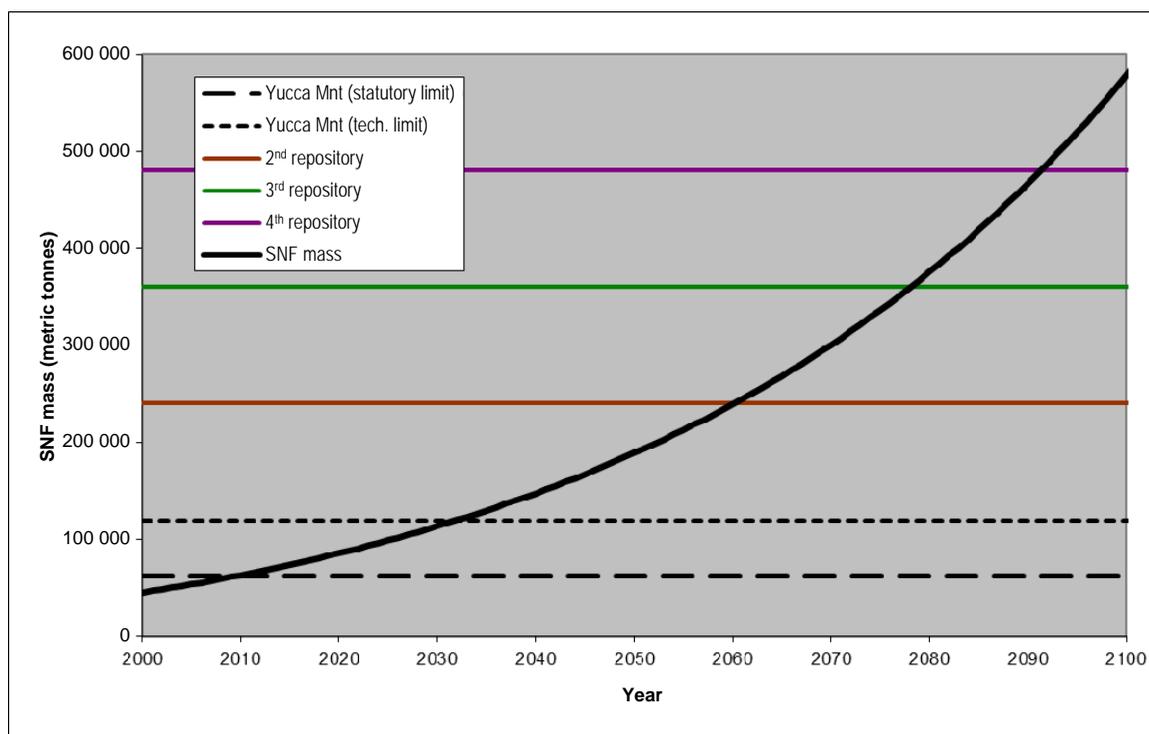
Spent nuclear fuel (SNF) reprocessing was pursued and performed on a small scale in the United States in the 1970s, then halted for almost 30 years for policy, but also economic, reasons. In the meantime, efforts have focused on managing the high-level waste (HLW) from the once-through fuel cycle. This approach converged in the proposed construction of the Yucca Mountain geologic repository. According to the US NRC website [1] “...on 3 June 2008, the US Department of Energy (DOE) submitted a license application to the US Nuclear Regulatory Commission (NRC), seeking authorisation to construct a deep geologic repository for disposal of high-level radioactive waste at Yucca Mountain, Nevada”.

While experts generally agree that this solution is technically sound, and would safely resolve the issue of all currently existing US commercial nuclear waste, it does raise some questions – it is not truly sustainable with the current once-through fuel cycle. Yucca Mountain repository would have filled up to its statutory limit around 2010-2014. Even though the actual technical limit may be about double and it would keep it open for another 20 or so years, a new such repository would still be required every several decades thereafter. The exact timing depends on the assumed repository capacity and electricity production growth, but an illustrative sequence is shown in Figure 1. (To generate Figure 1, it was assumed that the statutory limit for commercial SNF is 63 000 tHM, reached in 2010, actual capacity is 120 000 tHM, and subsequent repositories are also assumed to have 120 000 tHM capacity. Additionally, annual growth of 2% in nuclear energy generation was assumed.) Each time the SNF mass crosses a horizontal line, it indicates the need for another repository. It is difficult to envision adding multiple repositories at the required rate and demonstrating their performance for the next million(s) of years.

Already in the previous AFCI programme, fuel reprocessing had been reconsidered in the United States to reduce the repository load. With the latest US policy change with respect to used nuclear fuel management, DOE has been directed to examine a range of alternative solutions to Yucca Mountain, as demonstrated by the current DOE’s Fuel Cycle Research and Development Program (FCR&D Program) [2]. While instigated by different considerations, the approach proposed in this paper is highly compatible, and also based on understanding and addressing fundamental features and limitations of the nuclear fuel cycle.

## Objectives

To address the discussed concerns, this work proposes to set as the ultimate goal a nuclear fuel cycle that: i) generates HLW with “acceptable” isolation time requirement and radiotoxicity level; ii) “essentially” requires no permanent geological repositories. These top-level but vague objectives depend of course on the meaning of “acceptable” and “essential”. As the “acceptable” isolation time requirement, we propose to consider 300-500 years. The choice is somewhat

**Figure 1: Illustrative increase in SNF mass in the United States and the required repositories**


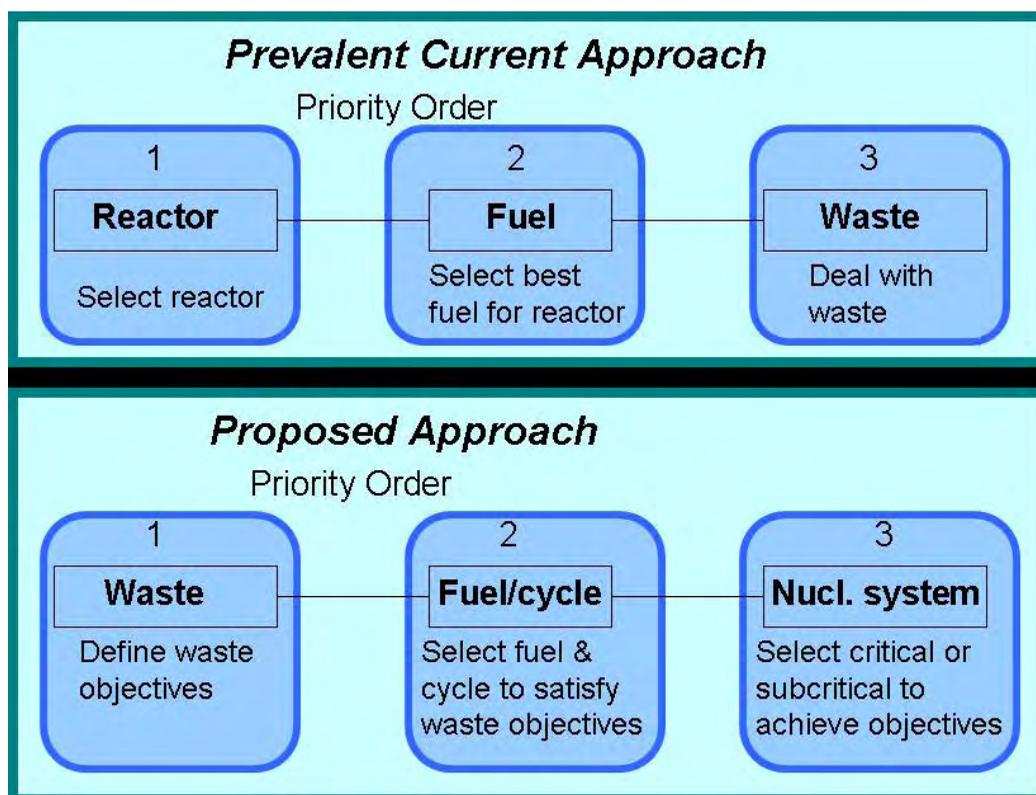
arbitrary, but defensible based on the possibility to reliably predict and ensure performance over such a period. The proposed “acceptable” radiotoxicity level is that which (after the isolation period) corresponds to or is lower than the equivalent amount of uranium ore that would have been needed in open cycle to produce the same amount of electricity. This criterion, while not as straightforward as it sounds, does provide a practical mean of comparing wastes without performing a specific risk assessment analysis. While some amount of HLW will be produced under any circumstances in any scenario, the second requirement is intended to indicate that no multiple large repositories would be needed.

## Metrics

The radiotoxicity was selected in the previous section as perhaps the most practical single parameter. For more realistic metrics, a compound weighted indicator may be used, accounting among others for the waste volume and mass, radioisotope activity, radiotoxicity, exposure paths and risk, economics, public acceptance, etc. This paper will mainly refer to radiotoxicity, without implying that this is the best factor, and allowing for future refinement of criteria to account for multiple indicators.

## Approach

It may be observed that traditionally a search for an effective fuel cycle solution would more likely than not proceed by selecting a specific reactor type, then the fuel form and finally searching for an optimum waste management solution within these bounding conditions. In this work we propose a comprehensive waste-management requirements-driven approach. In this approach, the search for a solution is driven from the opposite direction, i.e. from the fuel cycle back-end, starting with the waste management requirements, as indicated in Figure 2.

**Figure 2: Proposed approach, driven by the waste management requirements**

In this approach, the requirements (technological as well as stemming from public acceptance) imposed on the ultimate spent fuel or HLW stream are considered first, followed by the fuel form screening. Only then, the best mix of available nuclear systems and reprocessing satisfying these requirements is developed. There is no *a priori* bias for or against a given system (critical reactors, subcritical ADS, hybrid fusion-fission), fuel cycle (open, modified open, or closed), fuel type or form (U-Pu or Th-U, metal or oxide, TRISO particles), coolant (water, liquid metal, molten salt) and so on. The choice should be dictated by how well it satisfies the requirements. Naturally, limitations due to neutronics, safety, fuel performance, radiochemistry, economics and others have to be addressed and satisfied at the appropriate time.

One intended benefit is that a mix of several different systems will likely provide an optimum solution, promoting collaboration between the relevant industry and research entities in the fuel cycle back-end activities. The intent is also to avoid preconceived limitations and elimination, and keep all options open as long as meaningful.

Key attributes of the proposed approach are:

- *Public acceptance*: Addresses concerns on permanent disposal.
- *Goal-oriented*: Reduction of the waste to ore-level radiotoxicity in several hundred years.
- *Science-based*: Physics, engineering and economics dictates technology choices.
- *Solution-driven*: Each identified technology will be utilised in the most efficient way to reach the ultimate goal.
- *Unbiased*: All present fuel cycle technology (reactors, subcriticals, separation, fabrication) will be utilised to the optimum extent.
- *Forward-looking*: New technologies will be developed and introduced as necessary.

## Path to evaluation and implementation

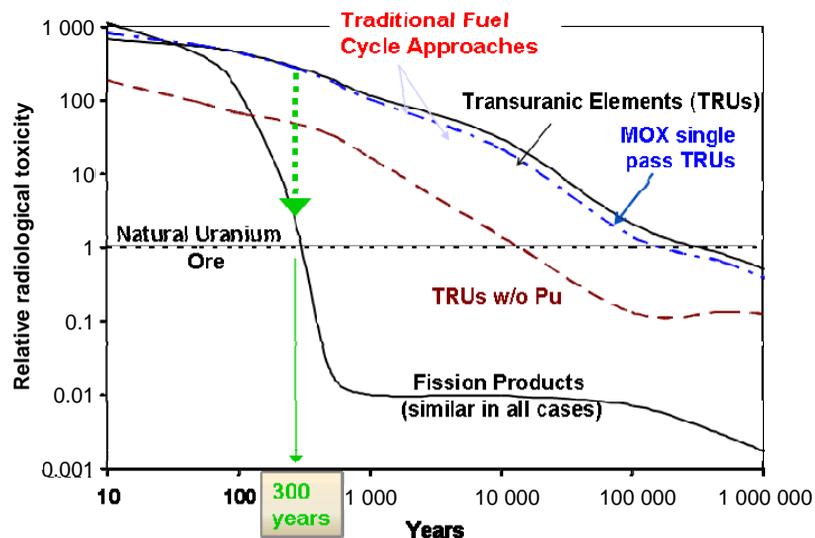
The envisioned path to evaluation and implementation includes the following stages and components: i) develop capability for extended scenario studies, including assessment of sustainability, radiotoxicity and proliferation resistance characteristics; ii) develop models of various reactor and reprocessing options; iii) develop isotopic mass balance flow sheets for various cases; iv) determine reactor and reprocessing specifications required to meet the 300-year waste requirement for each case, including the isotopic composition of fuel at reactor discharge, as well as isotopic composition of streams after reprocessing; v) identify technical, proliferation and cost issues for each case; vi) generate specifications for nuclear systems development programmes, including requirements on the fuel and system performance for the isotopes targeted for incineration; vii) generate specifications for reprocessing in terms of elemental recovery ratios.

## Basic considerations

The well-known relative radiotoxicity profiles for U-Pu cycle are shown in Figure 3. The once-through cycle takes between one hundred thousand and a million years, significantly longer than a few hundred years, to recede to the uranium ore level. Single plutonium recycle in MOX has only a very limited impact on radiotoxicity reduction. Complete recycle of plutonium has a significant effect, reducing the radiotoxicity by about one order of magnitude, but is still insufficient for reaching the objectives. The fission products decay below the uranium ore in about 300 years, which makes the 300-500 year objective for the overall radiotoxicity reduction practically coincident with being able to fully recycle all actinides until their final destruction from the recycled fuel. This strategy, besides reducing the volume and radiotoxicity of the waste, has the added advantage of a significant improvement of fuel utilisation. A number of conditions would need to be met, however, including:

- acceptable reprocessing recovery and losses fractions (key requirement);
- availability of adequate fast spectrum systems (critical, subcritical);
- feasibility of deep burn of radiotoxic actinides;
- fuel manufacturability and satisfactory performance;
- reliable and safe operation of all systems and facilities.

**Figure 3: Relative radiotoxicity for major fuel cycle and reprocessing alternatives**



It seems likely that a combination of fast spectrum burners will be required, in combination with ADS or hybrid fusion-fission systems to complete incineration. Two inherent challenges are present. Such multi-tier systems will lead to more complex design of reactors and subcriticals and to more demanding requirements on their operation, safety parameters and target design/performance. At the same time, the overall system would need to address both the “legacy” fuel (that will keep being produced for decades) as well as the “new” fuel(s).

Alternatively, Th-<sup>233</sup>U cycle, at least at the face value, offers potential to significantly reduce radiotoxicity and satisfy the waste management goals, due to significantly reduced build-up of higher actinides.

Finally, “revolutionary” systems and approaches are not excluded, as long as they offer potential to contribute towards reaching the waste management objectives.

## Reprocessing

Following the general approach philosophy, various reprocessing options will be examined, including PUREX, variants of UREX+, AIROX, pyroprocessing, Fluorex, Thorex, etc. The main discriminating factors leading the selection will be the radiotoxicity of the waste streams, proliferation resistance and economic effectiveness. It is possible that a tandem combination will be needed in order to meet the recovery fractions enabling the 300-year objective. The current state-of-the-art reprocessing technology could conceivably just meet these objectives for first recycle of LWR SNF. The increasing level of difficulty of recycled fuel, due to the higher MA content, activity and decay heat, will make it unlikely to meet these objectives with the available reprocessing technology.

## Thorium cycle considerations

Various specific features, advantages and disadvantages of thorium-bearing fuel and the Th-<sup>233</sup>U fuel cycle are discussed elsewhere. Here we briefly remind the reader of the thorium fuel advantage from the waste management standpoint. The interaction/transmutation chain including both <sup>232</sup>Th and <sup>238</sup>U, as well as transuranics, is shown in Figure 4. Due to its “lower” position, <sup>232</sup>Th requires multiple neutron captures to “progress” to TRU, which would under ideal conditions reduce the concentration of TRU (Pu+MA) by several orders of magnitude. The main issues related to thorium fuel include <sup>231</sup>Pa and the need for heavily shielded remote reprocessing.

Thorium has been considered for many decades and proposed by various organisations. Considering the front-end and core residence time only, it has not had much chance to compete economically with the U-Pu cycle. The reason is that the latter can rely on well-established industrial infrastructure and low price and availability. If the criterion is waste reduction/elimination, however, a Th-<sup>233</sup>U cycle (due to its significantly lower TRU/radiotoxicity) may become overall less costly, i.e. more economical. The radiotoxicity of the wastes from UOX and U-<sup>233</sup>Th recycle with different separation efficiencies is shown in Figure 5, showing the potential advantage of Th vs. U. This only illustrates thorium fuel potential and does not imply *a priori* selection of thorium cycle. The ultimate selection will be requirement-driven, as previously discussed.

## Progress to date

The progress to date may be summarised as follows. A novel waste requirement-driven approach has been developed. Establishing of simulation capabilities and analytic tools is either under way or completed. Core physics tools are being benchmarked or extended as needed. Preliminary calculations and benchmarks have been performed.

We have devised preliminary fuel cycle scenarios and are performing preliminary analyses to scope alternatives from the standpoint of satisfying the waste requirements, primarily expressed

through radiotoxicity evolution, but also accounting for economics and proliferation resistance. Specific analyses are being performed on thorium fuel implementation in multi-tier systems. The potential role of fission-fusion hybrids and ADS in multi-tier systems is considered for the next stage of analysis as well.

Figure 4: Chain depicting nuclear reactions and decay leading to build-up of higher actinides

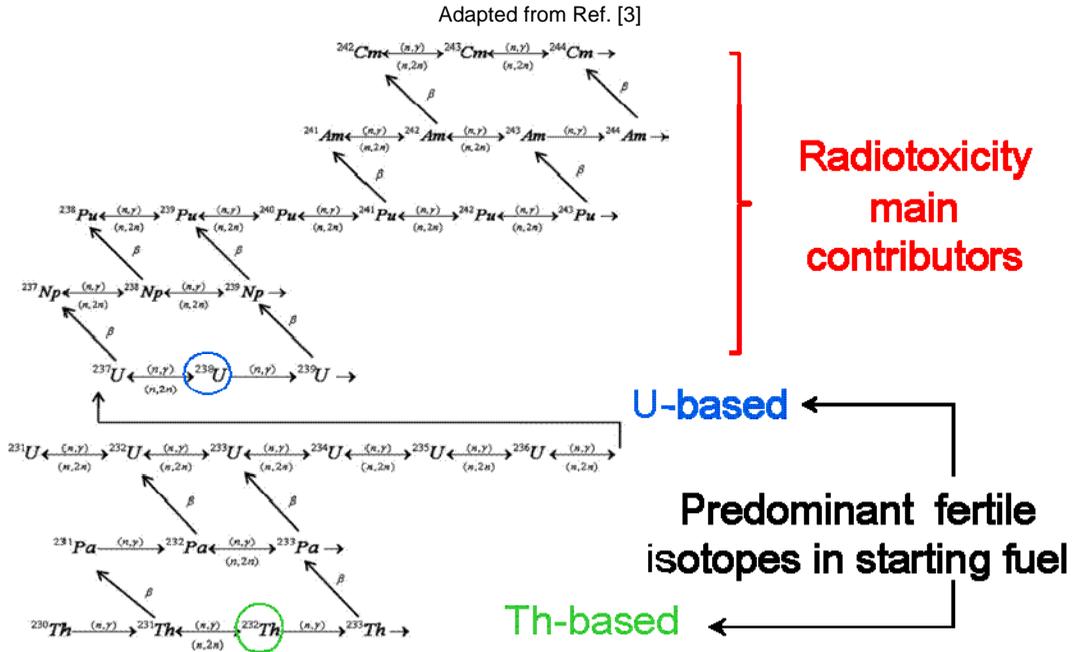
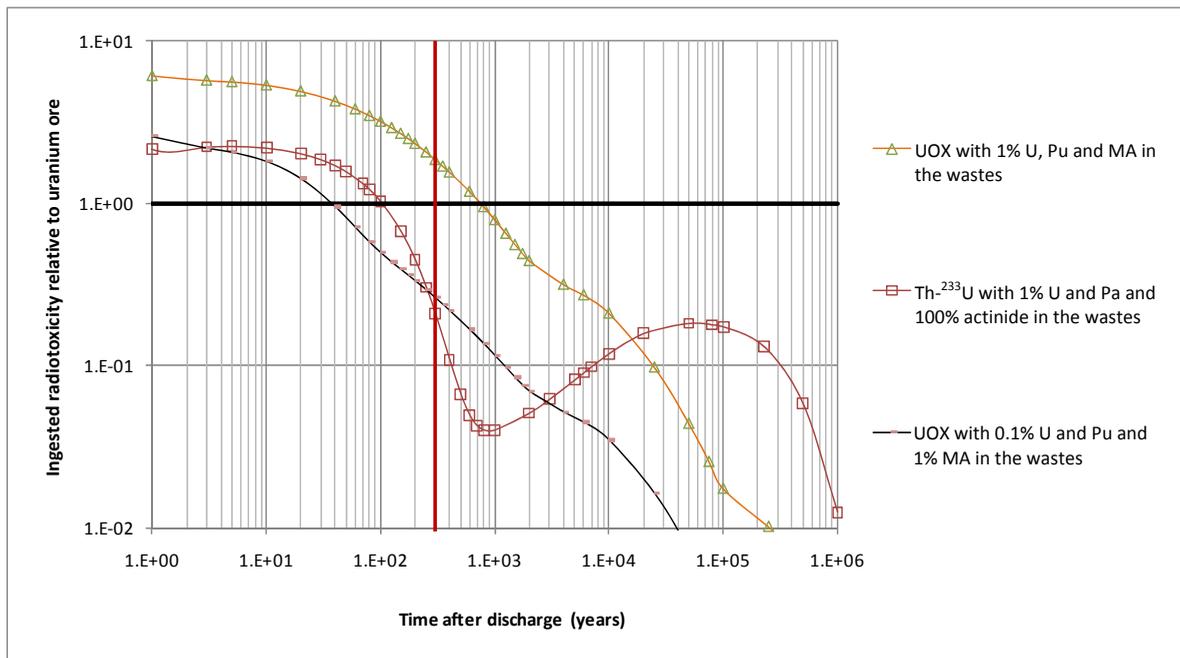


Figure 5: Illustrative waste radiotoxicity from UOX or Th-<sup>233</sup>U SNF recycle with different reprocessing efficiencies



## Summary and conclusions

A new approach to fuel cycle optimisation has been proposed. Rather than being based on the reactor type selection, it starts from and is driven by the waste management requirement, since this is one of the essential requirements for both the fuel cycle long-term sustainability and public acceptance. In this preliminary stage, tools for analysis are being established and exploratory analyses performed.

It is important to note that the methodology aims to eliminate any predetermined solution or *a priori* bias for a specific reactor or fuel. The decisions will be based solely on the match to requirements, and the spent fuel and high-level waste management will effectively guide the development of the fuel cycle and reactors. Consequently, these efforts are open to any technology that could contribute to accomplishing the stated goals.

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## World transition towards sustainable nuclear fuel cycle

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### Abstract

World energy demand will significantly grow in the mid-term future. It is foreseen that the world nuclear energy production share could drastically increase within the next century, according to many institutions' reports. Sustainable supply of these considerable energy needs by using only the thermal reactors is questionable due to a limited natural resource of uranium. Therefore the transition from the present open or partly closed to a fully closed fast reactor fuel cycle, implementing P&T for waste management and reusing fissionable material seems unavoidable in the mid to long term. The paper addresses these issues at the global level and discusses the transition scenario towards sustainable fast reactor fuel cycle using both a homogeneous (i.e. no distinction among different regions in the world) and heterogeneous (i.e. the world subdivided into different regions with e.g. different energy demand growth) transition scenarios. It will be shown how the need and the deployment pace of fast reactors in the different world regions is determined by the hypothesis of the energy demand growth, plutonium availability, natural uranium consumption (which, in our study should not exceed a critical limit), fast reactor core conversion ratio, ex-core time of spent nuclear fuel, target burn-up of UOX fuel impacting the quality of plutonium vector, and finally the reprocessing and fabrication capacities available in the future. Impact on high-level waste inventory to be disposed of and fuel cycle metrics such as radiotoxicity and heat load per energy produced will be assessed. Scenario simulations were performed with the COSI code developed by CEA.

## Introduction

The implications of a world transition from the current open or partially closed fuel cycles (FC) towards future sustainable closed fast reactor fuel cycle implementing partitioning and transmutation (P&T) still requires intensive investigations. Different deployment scenarios with various hypotheses on one side and extensive research and development (R&D) studies on industrial feasibility on the other are needed. Both the systematic analyses of possible transition strategies for each single country (or group of countries) and global scoping studies identifying key generic issues that impact strongly dynamic fuel cycle infrastructure must be performed. In order to co-ordinate efforts and to give the findings maximum credibility an international evaluation co-ordinating framework is important.

To support evaluation of R&D needs and relevant technology requirements, the NEA/OECD Expert Group on Fuel Cycle Transition Scenarios Studies was established. The tasks of the expert group are to assemble and to organise institutional, technical and economic information critical to the understanding of the issues involved in transitioning to long-term sustainable fuel cycle or a phase-out of the nuclear enterprise and to provide a framework for assessing specific national needs related to that transition.

The present study is more limited in scope with respect to IAEA INPRO studies (e.g. GAINS), and it is focused on a limited number of parameters in order to point out major trends and issues. In this respect, the use of thorium resources has not been considered, despite its potential and plans for future utilisation in some major countries (e.g. India).

## Scenario specification: General assumptions and hypotheses

In the first working phase two generic transition fuel cycle scenarios that best characterise the pathway envisaged by member countries were developed and a number of constraints was identified that may strongly influence the world transition. As a reference case a nominal homogeneous world treatment was considered in order to determine the global trend. Next, a heterogeneous world scenario study was assessed, using the subdivision into the four macro-regions encompassing countries with similar future economic development. An important aspect is to find the best assumption with possibly proper projection of energy production demands which provide a generic envelope for nuclear growth in each region and drive the scenario.

We have considered that a major parameter constraining the world transition scenario analysis is the available mass of natural uranium resources. One can argue that a better parameter would be uranium cost versus cumulative uranium production [1]. In fact, in that study it is argued that, since today uranium cost is ~2-4% of the cost of electricity, even a very sharp increase of cost to a sharp increase on uranium requirements for new reactors would not affect the nuclear electricity cost in a significant way. However, we think that, from one side, cost considerations and projections over a period of a century can be rather dubious and, on the other side, that one should pay more attention to practical and very important issues, as e.g. the pace of introduction of new mines in different regions in the world, with significant impact on the reliability of supply, that could modify and put in a very different perspective any hypothetical cost consideration.

As for natural uranium resources, according to *Uranium 2007* [2]<sup>1</sup> three cases can be distinguished for uranium resources: the first, without any upper limit including conventional,

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1. Recently published *Uranium 2009* added the high-cost category of <260 USD/kgU in response to both the overall uranium price market increase and increased mining costs. Considering the <130 USD/kgU class (highest class price in the 2007 publication), it reports a slight decrease of the identified resources, while considering the new added category cited above an increase of ~15% was registered. However, considering the total world uranium inventory (thus including also unconventional resources), a decrease of 0.76% or an increase of 1.57% is observed respectively taking into account the two categories (respectively, <130 USD/kgU and <260 USD/kgU); we can conclude that our results are not affected significantly by variations reported in the last NEA publication.

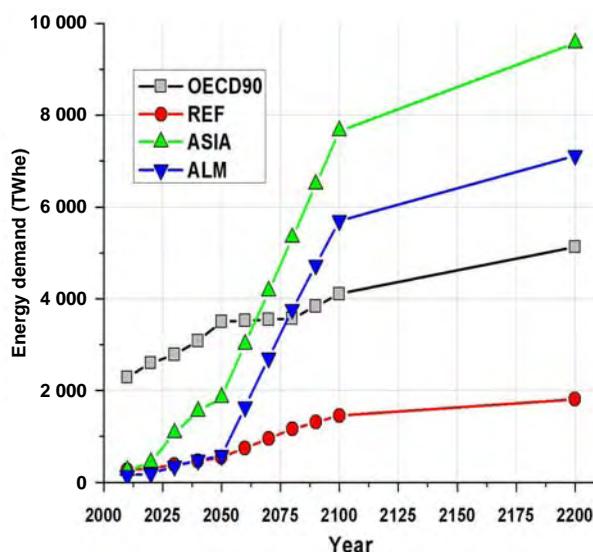
unconventional [3] and seawater uranium, the second with an upper limit ca. 38 MT (encompassing conventional and unconventional) and finally the third with a limit of ca. 16 MT, which refers only to conventional uranium.

The second key parameter driving the scenario is the growth rate of nuclear generation capacity. Table 1 and Figure 1 represent the data adopted for both the homogeneous and the heterogeneous cases.

**Table 1: Nuclear energy demands (TWh<sub>e</sub>): regional subdivision [7] rescaled to the B2-MiniCAM total value envelope [4, 5]**

	OECD90	REF	ASIA	ALM	WORLD
2010	2 289	268	252	145	<b>2 954</b>
2020	2 602	301	431	194	<b>3 528</b>
2030	2 780	383	1 072	348	<b>4 583</b>
2040	3 080	463	1 544	477	<b>5 565</b>
2050	3 502	542	1 846	580	<b>6 472</b>
2060	3 524	749	3 008	1 643	<b>8 926</b>
2070	3 547	955	4 170	2 706	<b>11 380</b>
2080	3 569	1 162	5 332	3 769	<b>13 833</b>
2090	3 836	1 307	6 494	4 733	<b>16 371</b>
2100	4 104	1 451	7 655	5 698	<b>18 908</b>
2200	5 130	1 814	9 569	7 122	<b>23 634</b>

**Figure 1: Nuclear energy demands (TWh<sub>e</sub>): regional subdivision [7] rescaled to the B2-MiniCAM total value envelope [4, 5]**



For the homogeneous case, the world nuclear energy envelope selected as a reference has been assumed to be in agreement with the data provided by the International Panel on Climate Change (IPCC) for one of the scenarios proposed to reduce greenhouse gas emissions by means of regional solutions (scenarios collected in the group called B2) [4, 5]. The scenario selected (namely “B2-MiniCAM”, by the name of the provisional code adopted to generate it) seems to reasonably reflect global nuclear energy developing trend up to 2100 reaching ~19 000 TWh<sub>e</sub> (more than 6 times than the present demand). It should be kept in mind in fact that in the near future there is the possibility that the world economic growth, and consequently the largest energy demand, will be focused in the most populated developing countries, and not necessarily in the OECD region [6].

Meanwhile, an additional hypothesis concerning the region-dependent energy demand evolution has to be assumed for the heterogeneous case. In general it was postulated that the subdivision proposed by the IPCC [5] in four macro-regions (namely OECD90: Western Europe, North America and Pacific, including Japan and New Zealand; REF: Russia, Eastern Europe and the former Soviet Republics; ASIA: Central and Pacific Asia, ALM: Middle East except Turkey, Africa, Latin America and the Caribbean) is sufficiently representative for world heterogeneous economies.

The regional subdivision proposed by the scenario selected (B2-MiniCAM) seems questionable (ASIA and ALM overtake OECD90 demand before 2035), therefore a more realistic subdivision proposed by the International Institute for Applied Systems Analysis (IIASA) (i.e. the “Middle Course” scenario, called B) [7] was adopted re-scaled preserving the B2-MiniCAM envelope trend line. In this way, the ASIA and ALM overtaking OECD90 date are shifted towards the end of the century (see Figure 1), and the total value remain reasonable (with a share of nuclear electricity production of about 25% in 2100). However, even if the hypothesis of energy demand growth in the ALM group of regions can still be considered questionable, it has the interest of showing areas where spectacular developments can be foreseen, obliging to go beyond “business as usual” studies, mostly performed for already well-developed countries.

For both the fuel cycle considered (homogeneous and heterogeneous), the total and regional energy envelopes have been adopted for the period 2010-2100, whereas in the subsequent period 2100-2200, the hypothesis of a slight increase, with a rate of 0.25%/year, has been assumed [4].

### ***Nominal case with “homogeneous world” approach***

As was indicated previously, in our study the major parameter constraining the world transition scenario analysis is the available mass of natural uranium resources. Consequently the reactor systems taken into consideration which impact the uranium consumption comprise three different reactor types: i) French PWR (N4 type) fuelled with UOX; ii) sodium-cooled fast reactor (SFR) with homogeneous core charged with mixed-oxide (MOX) fuel sub-assemblies containing minor actinides (MA) and with a conversion ratio (CR) close to one; iii) SFR Pu breeding systems loaded with metallic fuel (Zr supporting matrix containing transuranics fuel, TRU) with axial and radial blankets charged with uranium oxide, to improve the fuel breeding characteristics. All scenarios span over two centuries (from 2010 till 2200) and fast reactor deployment begins at 2050.

The flexibility of fast cores allows studying different conversion ratios like 1.0, 1.18, 1.47 or even 1.6. This last very high value is presently considered a valuable objective in countries, like e.g. India, which are looking for an early deployment of nuclear power to cope with their ongoing economic expansion.

As far as characteristics of fuel cycle are concerned it was assumed that the LWR spent fuel (SF) will be cooled five years whereas the fast reactor SF cooling time is case dependent and may vary within two or five years (SF sub-assemblies of blanket however will be cooled for two years). Another ex-core time such as reprocessing and fabrication time amounts both to 0.5 years. A common reactor load factor 0.85 was assumed for both thermal and fast systems. For fast reactor fuels a burn-up postulated was 136 and 85.6 GWd/tHM respectively for the isogenerator (CR = 1) and the breeder (CR = 1.47) systems. Inventory of spent fuel legacy in 2010 amounts to 157 560 tonnes for LWR systems, and 56 tonnes for fast reactor system (the isotopic composition has been determined by means of COSI simulation). Losses were assumed to be 0.1% for reprocessing and 0.2% for FR fuel fabrication. Plant thermal efficiency is 34% in the case of LWR, and 40% for FR.

### ***Regional subdivision with heterogeneous approach***

As for the energy demand, different hypotheses for each region concerning the fast reactor types and yearly start-up date have been assumed. In particular, OECD90 and REF deploy fast reactors (“isogenerators”, i.e. conversion ratio close to one) in 2040 while ASIA and ALM deploy high-performance breeder reactors, respectively starting from 2060 and 2080.

The objective of all scenarios is to tune the deployment strategy in such a way that fast reactors replace the thermal fleet in operation as soon as possible due to Pu availability and in order to save the natural uranium resource. Reprocessing capacity is to be kept constant in all regions up to 2030 (i.e. fixed to the level of capacity available at present) and then left as a free parameter.

### **Characteristics of reactor systems used in the scenario studies**

Following the hypothesis that either the breeder or isogenerator (i.e. self-sustaining) fast reactor technology could be ready for industrial deployment latest at 2050, this date was hypothesised as the beginning of the transition. For the world market two representative fuel cycles have been analysed for multi-recycling of transuranics by employing a sodium-cooled fast isogenerator (conversion ratio CR~1) and a sodium-cooled fast breeder system with higher conversion ratio (CR~1.5) and a reduced composite doubling time (CDT; it gives measure of the time needed to produce enough Pu mass for doubling, by means of identical reactors, the reactor fleet). The aim is assessing the transition period to optimise the material management, the resource consumption, and the fuel cycle infrastructures: reprocessing and fabrication capacities, including possible impact on high-level waste repositories.

In order to show the intrinsic flexibility of fast reactors in the subsequent scenario studies considered here, a wide range of core models has been assessed by means of the ERANOS code system [8]. For calculations, the JEF-2.2 nuclear data library has been employed [9]. The advanced burner reactor (ABR) [10] core has been considered as a reference for the models developed at KIT. By varying the fuel content and the geometry, fast cores with different intrinsic characteristics have been assessed. The reference ABR core is loaded with metallic fuel and has an internal breeding gain close to zero [10]. The Pu vector and MA content correspond to a typical LWR spent fuel composition with discharge burn-up of 50 MWd/kg and after a cooling time of five years (the MA/Pu ratio is ~0.1). Starting from that core, a breeder version has been assessed by adding radial and axial blankets composed of  $\text{UO}_2$  (99.75 wt.%  $^{238}\text{U}$ ) such that a very high conversion ratio (CR~1.5) has been achieved. The core model presents the same power density (MW/tonnes of Pu equivalent) as the Superphénix core and a composite doubling time of 17.8 years with in-pile fuel irradiation time of 1 200 days and ex-core lag time of 5 years (which, if is reduced to 2 years, allows a CDT = 11.7 years), resulting with an average burn-up of 85.6 GWd/t. The characteristics of the fast breeder system (metal core and oxide blankets) used for scenario analyses, presented in the following part of the paper, are shown in Table 2. Additionally an isogenerator (i.e. European fast reactor – EFR) characterised by a CR~1 was considered (characteristics are summarised in Table 2).

**Table 2: Characteristics of the breeder and isogenerator fast reactor systems considered**

	<b>Fast breeder core</b>	<b>Isogenerator core</b>
Core fuel type	(U-TRU)Zr	(U-TRU) $\text{O}_2$
Average Pu content (%)	17.25	22.21
Power (GWe)	0.56	1.45
Conversion ratio	~1.5	~1
Cycle length (EFPD)	300	340
No. cycles (core/blanket)	4/8	5/8
Fissile burn-up (GWd/tHM)	85.6	136

### **Nominal case study: World homogeneous transition scenario analysis and results**

Due to complexity of the world transition scenario advanced fuel, a dynamic fuel cycle analysis code, the COSI6 code (ver. 5.1.4) developed by CEA Cadarache [11], has been used for global assessments, tracking mass flow throughout the complete fuel cycle, including the front-end facilities (the mine, the enrichment and/or the reprocessing plants and the fabrication of fuel

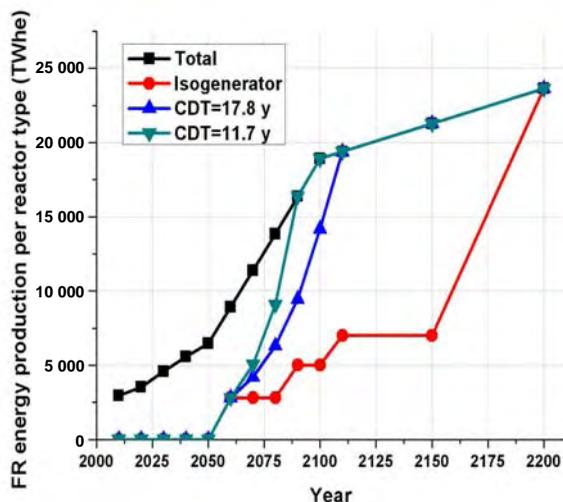
factories), the reactor, and the back-end installations, including the interim storage and the geological disposal. As indicated previously it was assumed that the transition starting date is 2050 and two closed fuel cycle schemes have been chosen: i) multi-recycling of TRU in a sodium-cooled fast spectrum isogenerator (CR~1); ii) multi-recycling of TRU in sodium-cooled fast breeder reactors (CR~1.5). The main goal is the assessment of the time at which the fast reactor's energy production will meet a given envelope of the world energy demand, and the consumption of natural uranium which should not exceed a hypothetical critical limit.

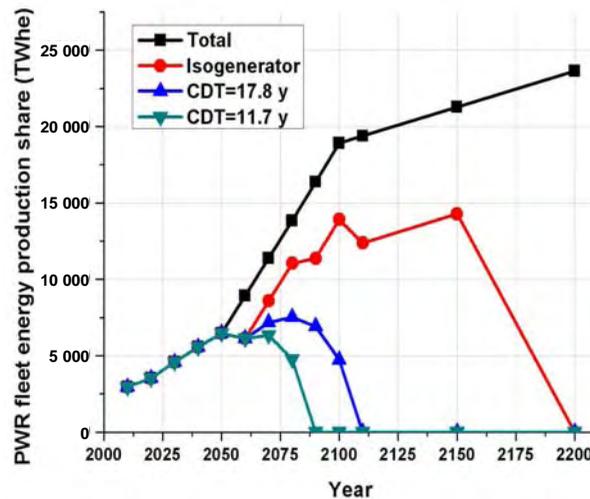
The nominal case scenario study represents a simplified reference approach whereby the world nuclear economy undergoes a unified treatment. Therefore it is assumed that all countries undertake a common effort deploying first thermal and then fast power plants. The use of isogenerator fast reactors is relevant for countries which have both a healthy nuclear economy (developing since the early 1970s) and a legacy stock of spent fuel containing plutonium to be reused. Transition scenario studies using this reactor type indicate that only a slow stepwise deployment schedule of isogenerator is possible due to the shortages in the fuel supply caused by the lack of separated plutonium for the fuel production. In fact, the available separated plutonium mass is a very sensitive parameter, depending on the fuel ex-core lag time (cooling and fabrication), inventory of spent fuels in interim storages, reprocessing capacity limitations, etc. The slow deployment pace is very probably consistent with the limited need in already developed countries but very probably not consistent with deployment needs in other regions, as will be shown later on. However, to cope with this difficulty, the use of fast reactors with breeding blankets, recycling their own fuel, allows more flexibility with respect to Pu management. This type of reactor (especially with dense e.g. metallic fuel) might be a preferable option for developing countries having extremely dynamical economic growth which will decide to go nuclear in the near future.

Transition using the fast breeder reactors significantly impacts the replacement scheme. Sensitivity analyses were performed on parameters such as composite doubling time and its effect on the deployment pace. Breeder reactors with a composite doubling time of ca. 18 and ca. 12 years were investigated. The results of the scenario studies are inter-compared in Figure 2 where the nuclear energy production share achievable with different breeder and isogenerator reactors is shown. The energy production of the world thermal fleet is given in Figure 3.

As far as fuel cycle characteristics, five years cooling period of discharged fuel and a one year period for reprocessing and fresh fuel fabrication were assumed (for spent fuel coming from both thermal and fast systems).

**Figure 2: Nuclear energy production share – fast reactor different options contribution**



**Figure 3: Nuclear energy production share – LWR different options contribution**

As expected, it is found that whatever the fast reactor type, the LWR remain a significant part of the system until the end of the century or even ten decades after the fast systems' introduction. The bottle neck is the inventory of TRU (especially Pu) required for fabrication of start-up cores which determines the rate of fast reactors' deployment. Fast reactors show a potential to cover fully the total energy demand: i) from year 2200 in the case the isogenerator fast reactors; ii) from year 2110 in case of breeder fast reactors with the composite doubling time of ca. 18 years; iii) from year 2090 in case of breeder fast reactors with the composite doubling time of ca. 12 years. Apart from CR, the key parameters having impact on the FR deployment pace are: the imposed energy growth rate (which exhibits in our assumptions a very steep slope in the time period from 2050 till 2100), the spent fuel inventory availability for reprocessing, the reprocessing capacity constrains (especially the imposed restriction to 3 800t/year until 2030) and the fuel cycle characteristics, and in particular the ex-core lag time.

As expected, different scenarios lead to different cumulative natural uranium consumption. As indicated in Figure 4, the scenarios adopting breeder reactors show significant reductions of consumed uranium mass. With respect to the LWR "once-through" fuel cycle [where the "once-through then out" (OTTO), fuel cycle is the reference for the analysis], the introduction of the fast systems enable to achieve an uranium resource saving in 2100 equal to 14% for isogenerator (CR~1), 37% for the breeders with CDT~18 y and 53% for the breeders with CDT~12 y (in 2200 the contribution of fast systems is respectively around 41%, 81% and 87%). In general the higher the conversion ratio the higher the rate of the fast reactor installed capacity, hence the more significant the uranium savings.

The required fuel reprocessing and fabrication needs are shown in Figures 5-10 for the transition towards fast reactors scenario. According to scenario hypotheses a gradual decrease of UOX reprocessing and fabrication capacities vs. time is observed together with an increasing of the FR share, which finally replaces completely the thermal fleet. In case of the two breeder options (with CDT~18 y and CDT~12 y), it is clear that FR facilities requirements follow tightly the considered energy demand, and that at the end of the present century their required capacities will be an order of magnitude higher than the present ones.

The impact of finite reactor lifetime and the start-up full core load on the consumed natural uranium resources has been investigated in case of transition from LWR fuel cycle towards fast breeder closed fuel cycle assuming 17.8 years composite doubling time. The calculations show an increase (with respect to the equilibrium case) of 3.7% of the uranium mass consumed if one takes into account the start-up cores in the simulations, a further increase of 5.6% when a 60-year LWR lifetime is considered and a 7.5% increase if a 40-year lifetime is considered.

Figure 4: Cumulative masses of consumed natural uranium – comparison among scenarios

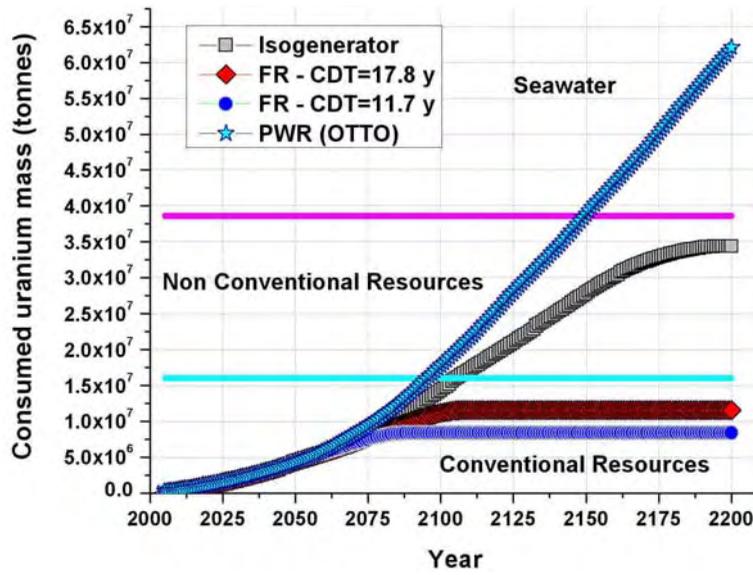


Figure 5: Reprocessing needs to manage isogenerators fast reactor fuel supply

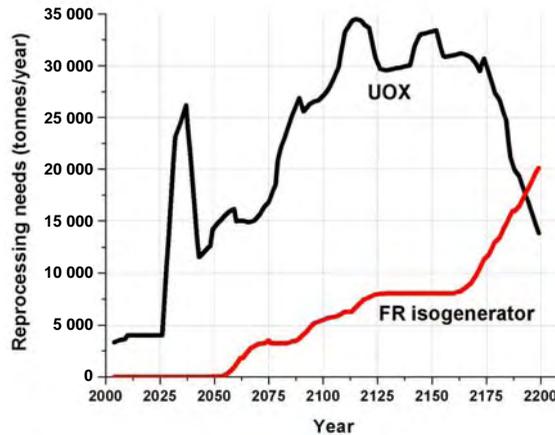


Figure 6: Fabrication annual capacity to manage isogenerators fast reactor fuel supply

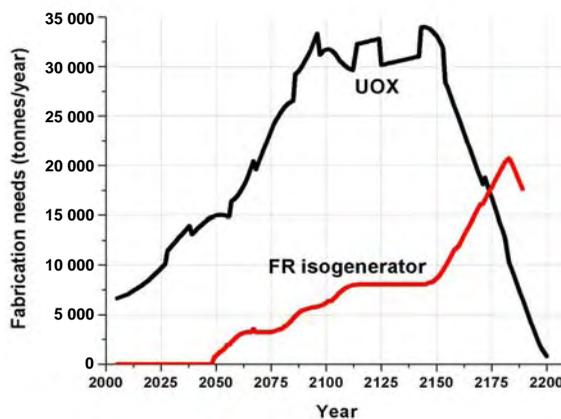


Figure 7: Reprocessing capacity to manage fast breeder reactor (CDT~18 y) fuel supply

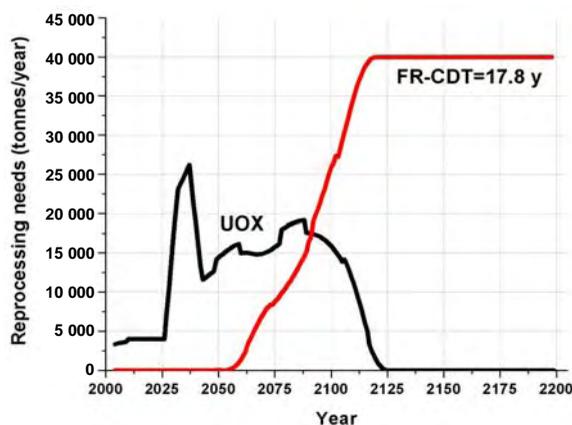


Figure 8: Fabrication capacity to manage fast breeder reactor (CDT~18 y) fuel supply

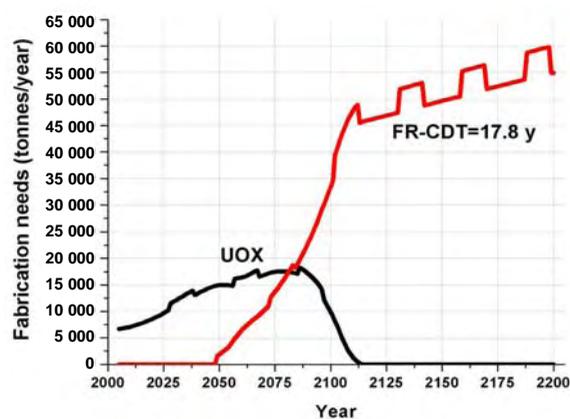
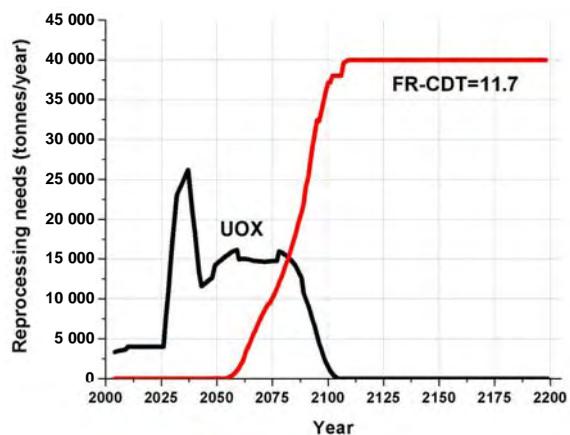
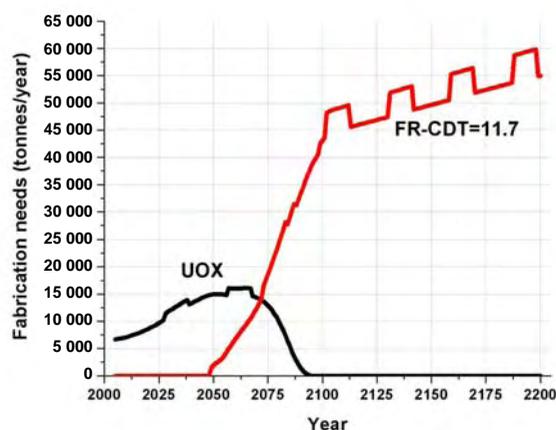


Figure 9: Reprocessing capacity to manage fast breeder reactor (CDT~12 y) fuel supply



**Figure 10: Fabrication capacity to manage fast breeder reactor (CDT~12 y) fuel supply**

### Regional approach

In the regional approach the transition towards fast reactors is based on the assumption that, as far as energy demand growth, the whole world can be roughly subdivided into four macro-regions characterised mainly by their economic growth factor [5, 12]. The energy production envelopes have been previously described and they have been applied to the regional analysis (Figure 1 and Table 1).

The assumed natural uranium resources are ~38.6 MT (value obtained by considering NEA analyses [2] and unconventional resources from [3]). The share by world region is roughly as follows: OECD90 59%, REF 11%, ASIA 5% and ALM 25%.

The spent fuel legacy was subdivided, due to the lack of published data, according to the nuclear energy production share of each region in the reference year 2000, resulting in a share for OECD90 83%, REF 10%, ASIA 6%, ALM 1%, respectively [13].

### Reference scenario with LWR deployment and open fuel cycle

If the “homogeneous” world nuclear energy requirement is met only by light water reactors (LWRs) the resource consumption and availability and spent fuel mass accumulation can be treated as a reference case. Natural uranium availability versus time is presented in Figure 11 and shows, around 2150, an exhaustion of uranium resource. Moreover, it can be seen that in 2100 the two curves, corresponding respectively to uranium residual availability and to the cumulative uranium consumption (in agreement with Figure 4), cross each other, indicating that half of the world reserves have been consumed. It is worth noting that, since the assumed energy demand at the end of the century will be roughly an order of magnitude higher than the present one, the residual uranium resources will run out in a relatively short period, as it will be shown. The uranium conventional resource limit will be hit in 2060, and 40 years later the unconventional resource limit will also be reached. These limiting dates would occur decades before if the engaged uranium had been taken into consideration.

Figures 12 and 13 show the uranium extracted mass (cumulative) vs. time and the yearly fuel fabrication capacity required for each region. The highest uranium demand comes from ASIA and, to a lesser extent, from OECD90. ALM, due to its steep increase in energy demand, will reach a value close to that of OECD90 in 2150. Finally, in 2150 REF, OECD90, ASIA and ALM will have consumed each 8%, 29%, 37% and 26% of the global available uranium resources, respectively. It is worthwhile, however, to mention that these numbers do not correspond to the actual uranium ore distribution in the world, and in particular ASIA will be forced to resort to consistent imports, owning only 5% of the global resources. The fuel fabrication facilities of ASIA and ALM require a

Figure 11: Uranium resource availability vs. time (case: only LWR)

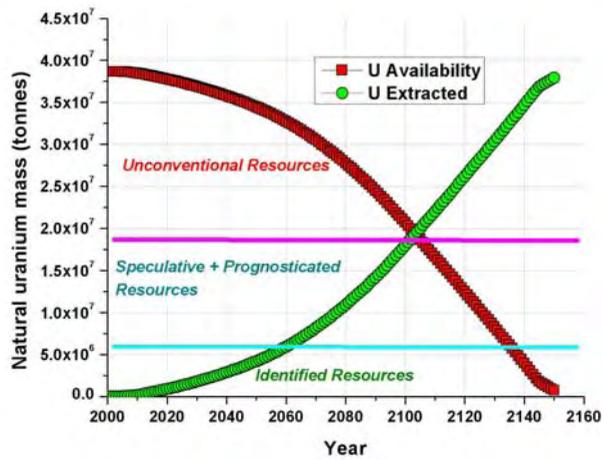


Figure 12: Cumulative masses of consumed natural uranium (case: only LWR)

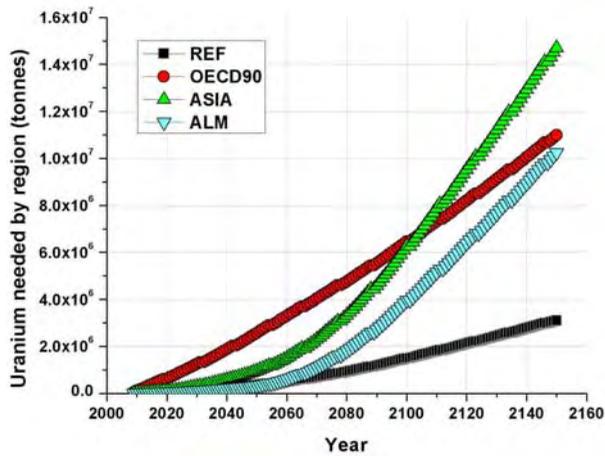
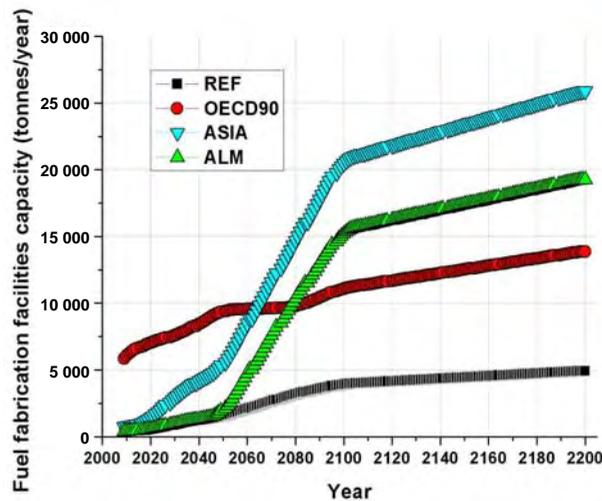


Figure 13: Regional fuel fabrication capacity (case: only LWR)

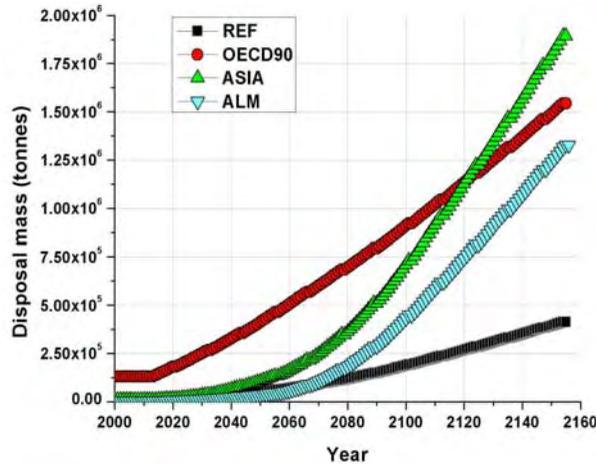


sharp capacity increase up to 2100; afterwards the increase of about 0.25% per year occurs following the energy demand slope [4]. The ratios of fuel fabrication capacities in 2100 with respect to 2010 are for REF, OECD90, ASIA and ALM 5.5, 1.8, 26 and 37, respectively. Figure 14 shows the spent fuel masses to be disposed of vs. time up to year 2150: ASIA produces the biggest amount of SF due to its high energy production, followed by OECD90 and ALM. Figures 15 and 16 show the spent fuel specific ingestion radiotoxicity (expressed in Sv/TWhe) and heat load (expressed in W/TWhe); as expected the regional behaviours are the same (because the same type of LWR has been considered for all regions).

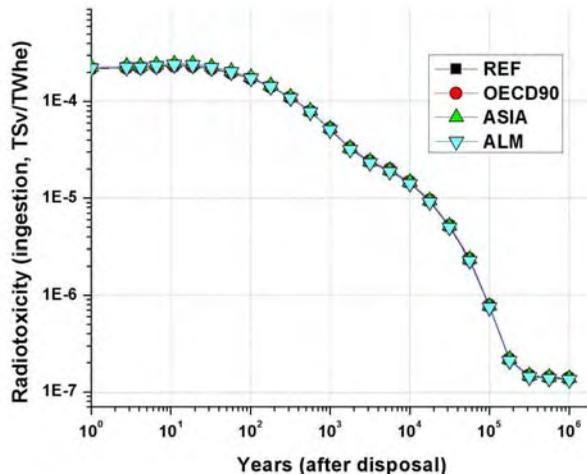
**World transition scenario towards fast systems in regional approach**

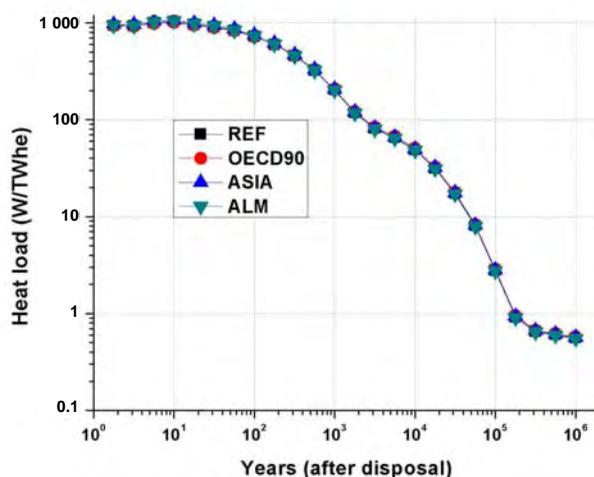
The transition from the regional scenario based only on the “once-through” fuel cycle to a fully closed fuel cycle (by the deployment of fast systems in all regions having a potential for such transition) requires a proper tuning of some key fuel cycle parameters. In particular, ex-core lag time driven by fuel cooling time impacts the composite doubling time and thus the deployment pace of FR. A two-year cooling time was chosen to impose a shorter fast reactor composite doubling time in the simulation, which is more realistic in particular for strongly developing regions.

**Figure 14: Spent fuel mass inventory per region (case: only LWR)**



**Figure 15: SF specific ingestion radiotoxicity evolution (case: only LWR)**



**Figure 16: SF specific heat load evolution (case: only LWR)**

In Figures 17-20 the total nuclear energy production is plotted for the four macro-regions, with the shares between LWR and FR fleets. As discussed previously, in ASIA and ALM, high performance breeder reactors (i.e. with CR~1.5 and relatively short composite doubling times, see above) were chosen for deployment due to high energy demand; their introduction was dated, respectively, at 2060 and 2080, while in OECD90 and REF isogenerators (i.e. CR~1, see above) were used, starting their deployment in 2040. The use of fast reactors with different conversion ratios in the different world regions is mostly driven by the pace of increase of the energy demand. Clearly in OECD countries, where a lower increase of energy demand is expected, the deployment of isogenerator fast reactors is probably the best choice, as also pointed out in Ref. [1].

However, that choice would be totally unjustified in countries that foresee a much more significant increase in energy demand. This fact points out the limitations of studies related to one specific country or to a group of countries, in particular if they have already reached a high level of energy production/person.

The pace of deployment was the fastest allowed by the Pu availability in the regional fuel cycle. The assessment shows that a complete shutdown of the thermal fleet is possible, at the latest, in 2130. This means that the transition to the new fuel cycle will last 50 to ~100 years, according to the hypotheses.

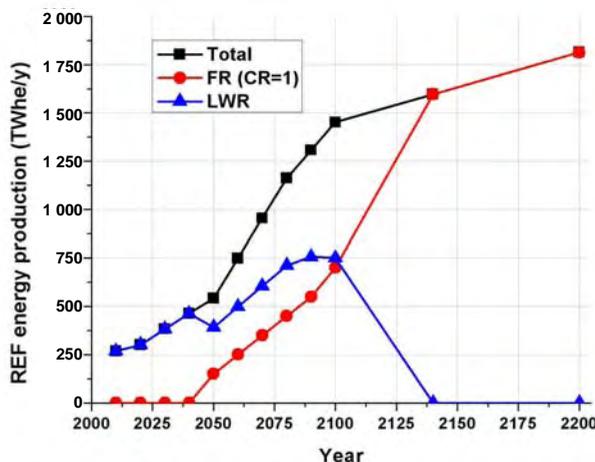
**Figure 17: Nuclear energy production share in REF (LWR and isogenerators)**

Figure 18: Nuclear energy production share in OECD90 (LWR and isogenerators)

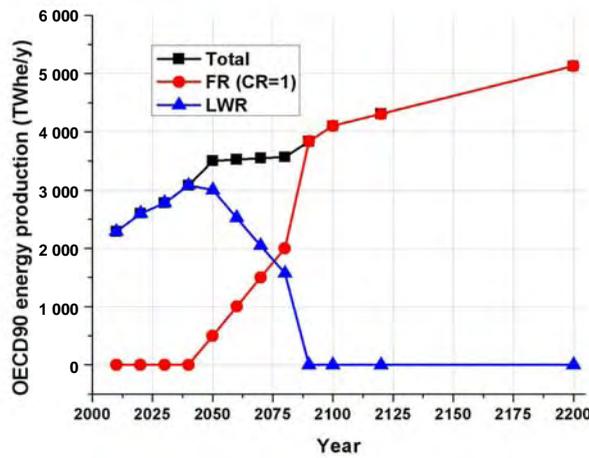


Figure 19: Nuclear energy production share in ASIA (LWR and breeders)

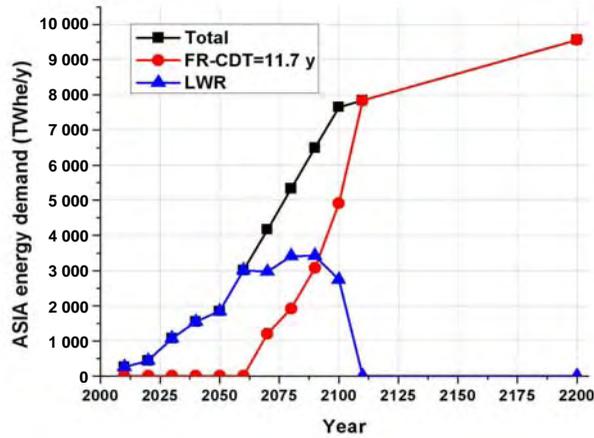


Figure 20: Nuclear energy production share in ALM (LWR and breeders)

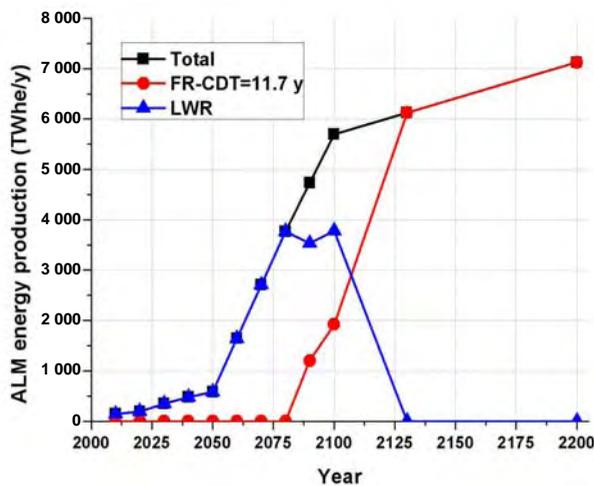
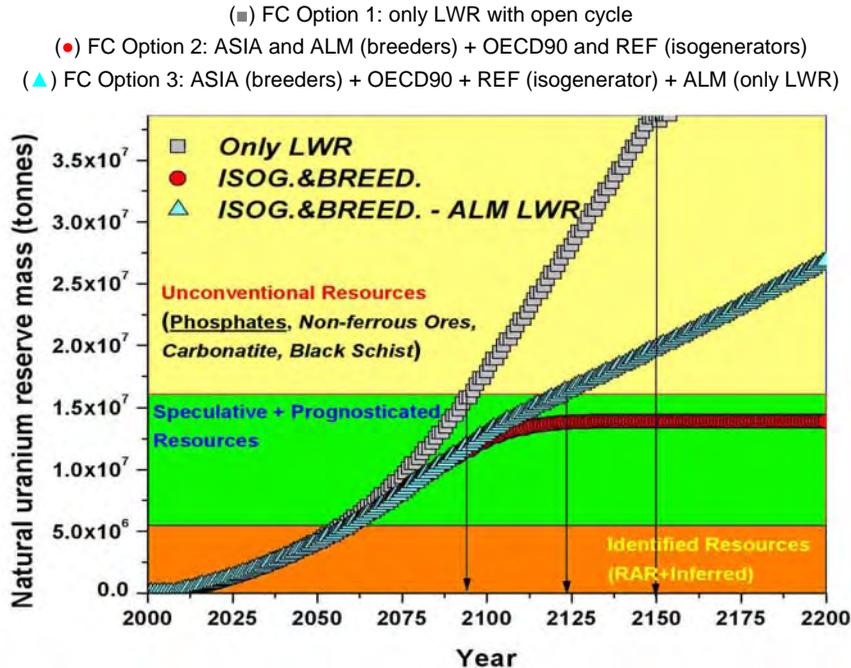


Figure 21 reports the variation of natural uranium resources with time for three FC options: i) the LWR “once-through” fuel cycle (in agreement with Figure 4); ii) the LWR + FR in all regions; iii) the LWR + FR in all regions except ALM where, in the hypothesis of a much more delayed deployment of FR, the nuclear energy needs are met only by LWR operating in an open cycle.

**Figure 21: Total natural uranium reserves for three different deployment scenarios**



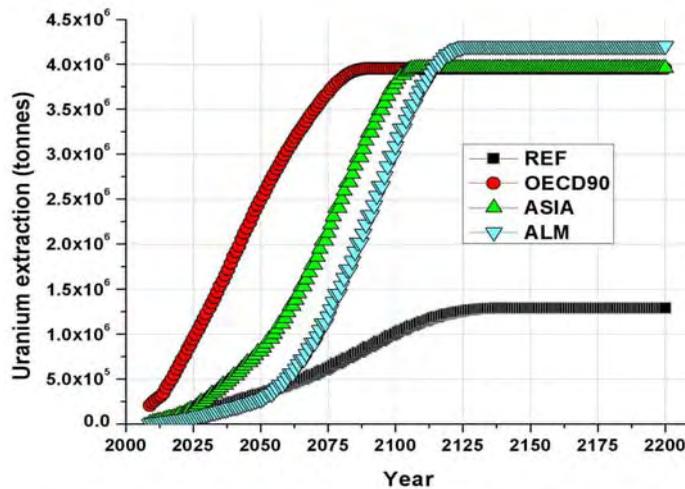
In the first FC option (once through), the natural uranium resources will run out at ~2150 and other types of uranium resources such as seawater should eventually be exploited, probably with a steep increase in the uranium price. For the second FC option (all regions adopt fast systems), the use of unconventional resources (i.e. phosphate rocks, carbonatite, black schist, lignite, etc.) will not be necessary, i.e. only mine resources will satisfy the global needs, although a significant effort for exploitation of new uranium deposits will be required. Finally, for the third FC option (all regions except ALM adopt fast system), starting in ~2120, the use of unconventional resources will be unavoidable.

More in detail, Figure 22 shows the uranium cumulative mass needed if all of the regions adopt fast systems (FC Option 2). ASIA and ALM will require very large amounts of uranium; the values are comparable (and even higher) than in OECD90. This is due to the hypothesis of a very aggressive policy of introduction of fast breeder reactor technologies with high breeding gain, short cooling times and early deployment. It should be pointed out that uranium mass consumed by ASIA is higher than that available in that region (5% of the total estimated resources).

The increase both of fuel fabrication and of reprocessing capacity, as required in this scenario study is one of the most significant results of the present analysis. A much accelerated pace of growth of the different installations is obviously related to the hypotheses made on new reactors' implementation. For example, with respect to the present LWR UOX required fabrication capacity (~9 000 tonnes/year), the COSI simulation indicates that in ASIA and ALM a UOX fabrication capacity of ~10 000 tonnes by ~2057 or ~2070, respectively, would be required, while in the OECD countries that capacity should reach ~11 000 tonnes by ~2040. Due to FR implementation strategy envisaged in the scenarios, the UOX fabrication capacity requirement will decrease after a few decades and a sharp increase of the FR fuel fabrication should be

expected: ~5 000 tonnes in the OECD countries by ~2080; ~18 000 tonnes in ASIA by ~2110 and ~14 000 tonnes by ~2130 in the ALM group of countries.

**Figure 22: Cumulative masses of consumed natural uranium (case: FR in all regions)**



As for the annual reprocessing capacity as it exists today (mostly in the OECD countries), i.e. ~3 800 tonnes, a value 6-7 times higher should be expected in ASIA by ~2100 and a value 4-5 times higher in ALM by ~2130 while an increase by a factor ~2-3 should be expected by ~2050 in the OECD countries. In practice, this would mean that the ALM and ASIA reprocessing capacities should be increased by about 2 500 tonnes every 10 years (equivalent to a La Hague size plant every 7 years).

These very large numbers should be taken as a warning of the potential infrastructure growth problems that could be faced in the ASIA and ALM groups of regions. Even if these difficulties could suggest a more realistic deployment pace, the challenges facing the different world regions are clearly pointed out by the present scenario study.

In Figures 23 and 24 the specific ingestion radiotoxicity (Sv/TWhe) and heat load (W/TWhe) evolutions in the waste disposal are shown for each region (FC option: FR in all regions). For the greater part of the time the highest level is that of ALM (due to later introduction of FR technologies); this trend is inverted after 200 000 years but differences between the four macro-regions are however not great.

**Figure 23: SF specific ingestion radiotoxicity evolution (case: FR in all regions)**

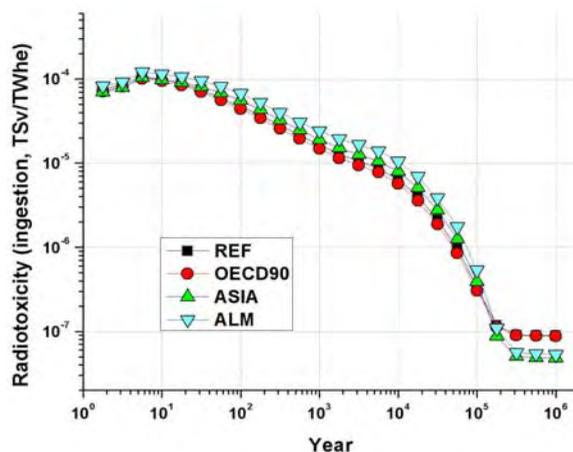
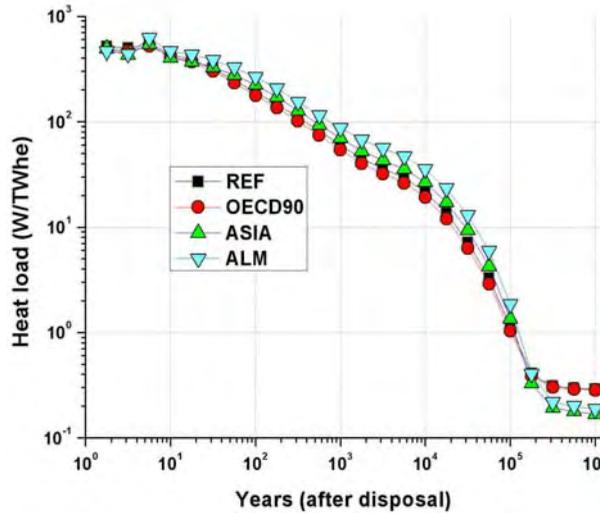
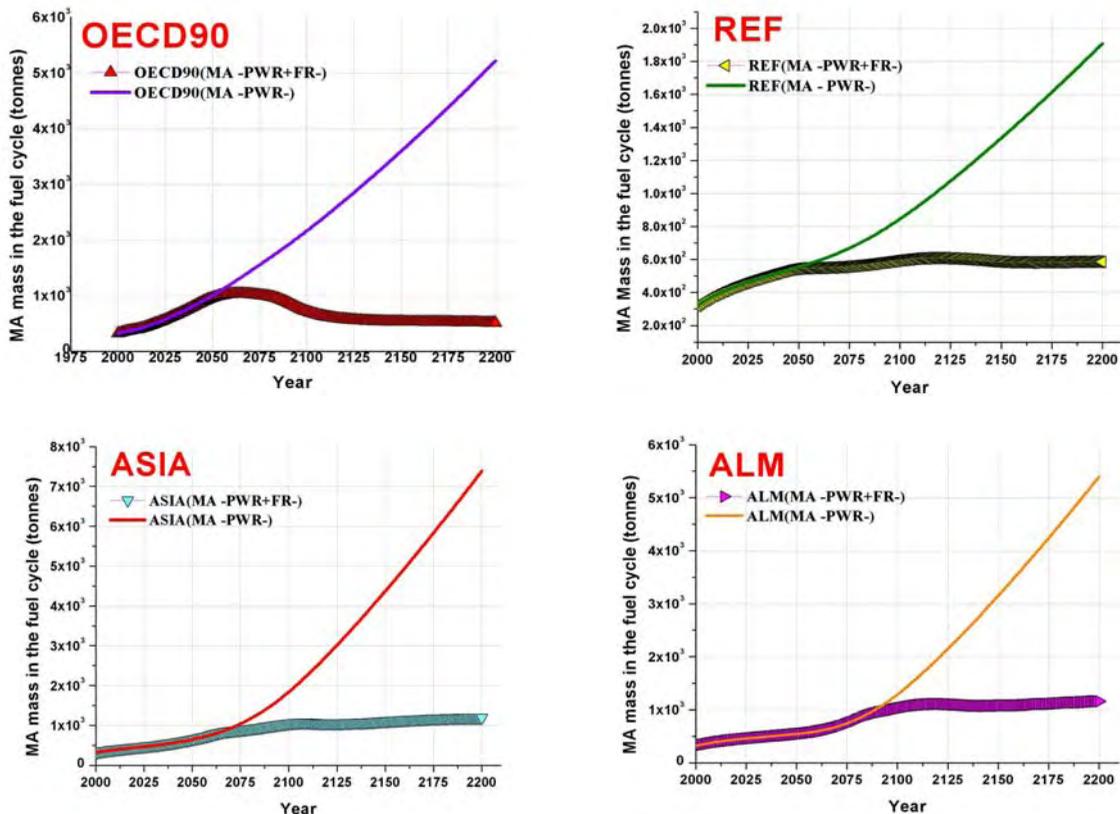


Figure 24: SF specific heat load evolution (case: FR in all regions)



Finally, in Figure 25 the mass evolution of MA (Am+Np+Cm) for the LWR “once-through” scenario (FC Option 1) and for the scenario with LWR and fast systems in all regions (FC Option 2) is compared. The bell shape trend in case of OECD90 is given by the assumed energy demand (roughly constant between 2050 and 2080, see Figure 18) and the contemporary introduction of FR for energy production. It was found that the transition towards FR will allow in some cases to reduce the mass of the MA produced from a minimum value of 3 (in REF) to 10 (in OECD90).

Figure 25: Comparison of the minor actinide (Am+Np+Cm) inventory between FC Option 1 (only LWR) and FC Option 2 (FR installed in each region)



## Conclusions

A simulation of a world transition scenario towards possible future fuel cycles with fast reactors was performed using both a homogeneous and a heterogeneous approach for the different regions. The heterogeneous approach considered a subdivision of the world into four main macro-regions (where countries have been grouped together according to their economic development dynamics). An original global electricity production envelope was used in simulations, and a specific regional energy share was defined. In the regional approach two different fuel cycles were analysed, the reference once-through LWR cycle, and a transition to fast reactor closed cycle for a better management of resources and minimisation of wastes.

In this respect, we considered that the potential future scarcity of uranium resources is not at all unreasonable, but a very serious perspective for the regions of the world where the energy demand growth is and will very probably continue to be spectacular.

The main objective in both cases (homogeneous and heterogeneous world approaches) was to deploy fast units as quickly as possible and to replace the thermal reactor fleet by the fast reactor one in order to minimise the uranium resource consumption and to cope with steeply increasing global world energy demand.

The results of this study are obviously very much related to the hypotheses made, in particular in terms of energy demand growth. However, some general trends seem to be of a general value, and can motivate further studies.

It was confirmed in this investigation that a rapid development of fast reactors, especially in the areas with expanding economies and strong energy demand growth, is essential for FR sustainability, for global saving of natural uranium resources and for reducing high-level waste generation which has to be disposed of.

In the case of an open cycle an increasing pressure on the uranium market and consequently on the price of uranium ore should be expected no later than at the end of the current century. Moreover, the increase in mining needs of unequally distributed resources can be a factor of uncertainty with an impact potentially more important than uranium cost considerations.

It would be, however, a very significant challenge to develop suitable fuel cycle infrastructure especially in the world regions that presently have limited (or no) nuclear power plants. In fact, the needed fuel fabrication and spent fuel reprocessing capacities should increase by at least one order of magnitude.

Fuel cycle facilities for uranium extraction, enrichment fabrication, reprocessing and storage of spent fuel and retrieved fissionable material must be technologically feasible and successively built in order to efficiently manage the greatly increasing fuel supply required for rapid transition.

Use of fast breeder reactors is indispensable if one tries to provide a global world perspective; their composite doubling time represents a major key parameter determining the deployment pace.

Of course, in well-developed regions of the world, where a more modest increase of the energy demand is expected, the deployment of fast reactors and its starting date are more debatable, as it is the assessment of an optimum value of the conversion ratio for these reactors and their potential contribution to waste management and minimisation.

The support of the thermal reactor fleet in the mix will in any case be needed until the end of the present century and even beyond, depending on the reactor type and the global or regional Pu mass availability.

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## Overview on homogeneous and heterogeneous transmutation in a new French SFR: Reactor and fuel cycle impact

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### Abstract

*In the framework of the 2006 French act relative to the sustainable management of radioactive materials and waste, CEA, AREVA and EDF are performing a collaborative development work on sodium-cooled fast reactor technology. This law also indicates the need, in 2012, of an assessment of the industrial outlook path and to put into operation a prototype reactor by 2020. The French SFR prototype ASTRID takes place in the context of this roadmap.*

*This paper presents an overview of the different options suitable from the technical point of view for minor actinide (MA) transmutation and their associated management in a fast reactor: homogeneous and heterogeneous modes with multiple recycling. Both methods have merits and drawbacks, depending on the reactor and/or cycle point of view. This study reviews, on the whole fuel cycle, from fabrication to partitioning and passing by reactor operation, the impact of transmutation for the two selected transmutation methods, taking into account all MA with or without curium. The comparison is based on a French Generation IV 3 600-MWth sodium-cooled fast reactor design with MOX fuel.*

*For the homogeneous method, a multirecycling of MA together with plutonium is studied, and for the heterogeneous method the minor actinide bearing blanket (MABB) with depleted uranium oxide matrix is selected.*

## Introduction

The scientific feasibility of long-lived waste transmutation has been studied for several years by the scientific community (including at CEA). In France transmutation in nuclear reactor has been studied since 1991 in the framework of the first French act relative to sustainable management of radioactive materials and waste. This first stage was closed in 2006 after 15 years of research. The main conclusions for the transmutation point of view were:

- Long-lived fission product transmutation is not efficient in nuclear reactor.
- Transmutation is preferable in a fast spectrum, and is not efficient in PWR.
- A global scenario approach is needed to evaluate all impacts.

In 2006 a new act was voted by the French parliament. To ensure accordance with the principles set out in environmental regulation, management of long-lived radioactive waste with high or medium activity, research and studies relating to such wastes are prosecuted under the three following complementary areas:

- 1) The separation and transmutation of long-lived radioactive elements. The corresponding studies and research are being conducted in conjunction with those carried out on new generations of nuclear reactors to dispose of, in 2012, an assessment of industrial perspectives and to operate a prototype facility before 31 December 2020.
- 2) The reversible disposal in deep geological formations. The corresponding studies and research are being conducted to choose a site and design a storage facility so that, given the results of studies conducted, the request for authorisation can be considered in 2015 and, subject to this authorisation, the disposal centre can begin operation in 2025.
- 3) Interim storage. The corresponding studies and research are being conducted for, by 2015, creating new storage facilities or modifying existing facilities to meet needs, particularly in terms of capacity and duration.

To meet the demands formulated in the first axis concerning transmutation, CEA, AREVA NP and EDF R&D have conducted assessments on the impact of transmutation on the new sodium fast reactor design (SFR). The design of the next generation of nuclear reactors is strongly correlated to their ability to deal with Generation IV goals: sustainability (including the minimisation of nuclear waste and the reduction of the long-term stewardship burden), improved safety and reliability, economic competitiveness, proliferation resistance and physical protection.

## SFR design

Current design studies on sodium fast reactors (SFR) [1] differ from those performed in the past in that the design criteria are now those of Generation IV reactors. This means that the core should have improved safety feedback coefficients (sodium void-coefficient, Doppler worth...). The innovative design is based on an oxide fuel pin technology to benefit from important feedback of past experience, and the core is at least break even.

The first pre-conceptual design phase is currently achieving a rather detailed SFR core design. Multi-disciplinary studies were performed at different scales: fuel pin, fuel assembly and core. Different options were evaluated in terms of neutronic performances and also potential gains of safety [2, 3].

The reference SFR core design is based on oxide fuel and on the following specifications:

- core diameter: < 5 m;
- core shape H/D ratio: > 0.2;
- maximal cladding dose: 200 dpa;

- maximal burn-up: 150 GWd/tHM;
- maximal linear power (at 15 at.%): 420 W/cm;
- sodium void reactivity: < 6\$;
- internal breeding gain ratio (IBG): > -0.05.

The plutonium isotopic composition is indicated in Table 1, which corresponds to the average plutonium isotopic vector available in France in 2035. The main core characteristics are given in Table 2.

**Table 1: Isotopic composition of “Pu 2035” in atom%**

<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>240</sup> Pu	<sup>241</sup> Pu	<sup>242</sup> Pu	<sup>241</sup> Am
3.6%	47.4%	29.7%	8.2%	10.4%	0.78%

**Table 2: Advanced SFR core characteristics (SFR V2)**

	Advanced SFR core
Core power (MWth)	3 600
Core electric power (MWe)	1 500
Core batches	5
Mass of Pu (t)	12.5
Mass of equivalent Pu (t)	8.1
Mass of U+Pu+Am (t)	74
Average Pu mass content (%vol.)	15.8
Core diameter (m)	4.9
Core height (m)	1
Core volume (m <sup>3</sup> )	17.4
Fuel residence time (EFPD)	2 050
Average burn-up (GWd/t)	99
Maximum burn-up (GWd/t)	139
Maximum neutron dose (dpa)	148
Power density (W/cm <sup>3</sup> )	207
Maximum linear power (W/cm)	420
Sodium void reactivity (\$)	4.9
Internal breeding gain	+0.04

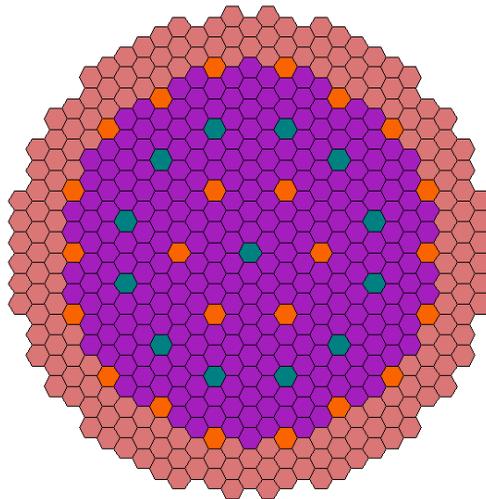
The core layout is presented in Figure 1. Even if not shown, the possibility of having one row of radial blankets is taken into account.

### Transmutation of minor actinides

The principle of transmutation consists in recycling waste in the nuclear reactors while seeking MA to favour their fission (“final destruction”), compared to the neutron capture (which does nothing but moves the problem...).

From the physics point of view the transmutation potential of a nuclear reactor could be evaluated firstly by the examination of the cross-section to determinate the way of interest. It is also necessary to consider the neutron potential in order to be able to quantify the number of neutrons available for transmutation (see Table 3). This approach (D-method) was described in Ref. [4] and allows assessing the capability to work at critical conditions and to produce the excess of neutrons for transmutation. It is based on the probabilistic approach applied to the cores at the “equilibrium” state.

**Figure 1: SFR core layout**



**Table 3: D-values for MA in thermal and fast spectrum**

	Thermal spectrum	Fast spectrum
<sup>237</sup> Np	-1.4	+0.3
<sup>241</sup> Am	-1.4	+0.3
<sup>243</sup> Am	-0.7	+0.3
<sup>243</sup> Cm	+1.6	+1.8
<sup>244</sup> Cm	+0.4	+1.1
<sup>245</sup> Cm	+2.0	+2.2
<b>AM</b>	-1.2	+0.4
<b>Pu + AM</b>	-0.2	+0.9

The advantage of the fast spectrum is easy to discern, because all the MA give a positive neutron balance. To summarise, we can say that fast reactors (FR) allow great flexibility in comparison with thermal reactors.

The transmutation of MA can be achieved with various modes of transmutation and waste management in FR.

Two methods of transmutation:

- The homogeneous mode whereby the minor actinides to be transmuted are directly mixed in the driver 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 of 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 into the following cycle. In this way, only reprocessing losses go to the ultimate waste.
- The once-through method: In this case the minor actinides are transmuted in targets where very high burn-up is reached.

In order to reach significant transmutation rates and to offer the capability to manage a large amount of MA (at the nuclear fleet scale) the multirecycling of MA is considered.

### ***Homogeneous method with multirecycling in SFR***

Several studies showed the physical feasibility of actinide multirecycling (Np, Am, Cm) together or separated in a fast reactor [5].

The limitation for actinide loading is due to the negative impact on safety parameters like sodium void and Doppler effects. The allowed limit, less than 5%, is well below the quantities necessary to equilibrate the reactor fleet. But it could be close depending on the need of MA to transmute in a scenario approach (for example in the case of a transient scenario between PWR and SFR).

The homogeneous method and its management have been deeply studied in the past. Here, actinides are reprocessed in the standard spent fuel flow, mixed homogeneously with plutonium during fresh fuel fabrication and re-loaded in the core maintaining a closed fuel cycle. In recent years, several studies [6, 7] have been performed at CEA considering sodium-cooled fast reactors (SFR). For many reasons the minor actinide part of the fuel cannot reach more than few per cent:

- Safety considerations: The introduction of minor actinides worsens the sodium void effect, the Doppler effect, the delayed neutron fraction and lowers the MOX melting temperature.
- Core design considerations: An increase in the helium production during irradiation may impact the S/A design (gas expansion volumes and pellet size to be reconsidered).
- Fuel cycle considerations:
  - Transportation, storage, and handling are affected by the presence of S/A with higher heat levels than traditional MOX S/A.
  - In a closed cycle strategy all manufacturing and reprocessing plants have to be adapted to deal with high equivalent doses, neutron source and thermal power levels.

### ***Heterogeneous method with multirecycling in SFR***

An alternative method consists in decoupled fuel/actinide management. In this framework, several options can be investigated. In the 1990s many studies were performed at CEA, focused on designing specific targets based on inert matrix than could be able to reach high transmutation and fission rates (“once-through” option).

In recent years, new studies on the heterogeneous solution – based on depleted UO<sub>2</sub> radial blankets with a significant amount of MA – have shown promising performances for this option. The major advantages of this totally decoupled management are the following:

- This option has only a small impact on safety coefficients (few %).
- It has almost no impact on the core management.
- It enables to optimise specific S/A in order to reach good transmutation rates.
- It separates the fuel cycle heavy nuclide streams (especially the front end) between electricity production and transmutation goals; the MOX fuel for the SFR fissile cores is not disturbed by MA and MA are concentrated in a reduced stream of heavy nuclides. In case of fresh fuel fabrication for the heterogeneous transmutation solution a dedicated facility is necessary in addition to the standard facility for the MOX driver fuel.

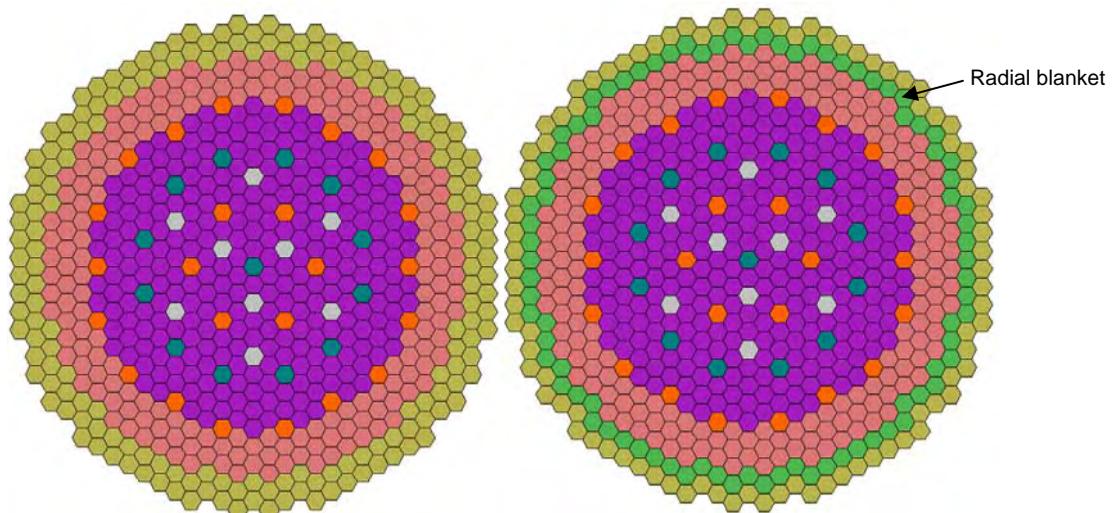
Despite all these advantages, this concept faces sharp R&D challenges on many fields that have to be carefully considered for the assembly design: helium production, fuel behaviour under irradiation, decay heat level, neutron source, fresh fuel thermal power, reactor operation, etc.

Between the homogeneous mode and the fully decoupled heterogeneous mode exists a range of intermediate solutions in which minor actinides can be put in some specific areas of the fissile core: those solutions can be classified as both homogeneous because MA are mixed with

the driver fuel and heterogeneous because only some fraction of the driver fuel assemblies are involved. A criterion that drives the definition and choice of these solutions is connected to the corresponding impact on safety parameters such as sodium voiding effect and Doppler effect. In that respect, the external core region in which the flux gradient is important appears as an interesting place to investigate, in order to limit the impact on safety-related core parameters keeping acceptable neutron fluence [8].

A huge amount of R&D is necessary in order to be able to conclude or not on the industrial feasibility of manufacturing transport and reprocessing issues.

**Figure 2: Core layouts without and with radial blanket**



### **Scenario approach**

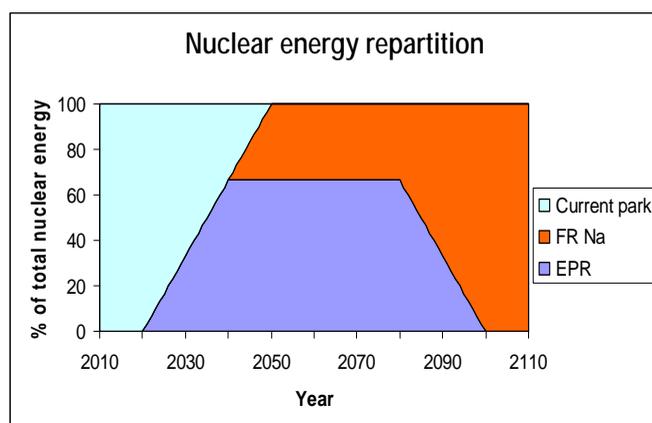
The aim of this section is not to specifically describe why and how the scenarios studied are performed, but rather to emphasise the need to perform this kind of study in order to determine the most consistent results in a comprehensive approach.

CEA is studying scenarios of principle for the French case through a dynamic vision of nuclear development, based on the one hand on the solutions offered by the existing fleet (reprocessing, manufacturing and NPP) and on the other hand on the solutions offered by the future reactors of fourth generation. This approach allows to estimate the global impact on the whole nuclear cycle and to evaluate the maximum values of required capacities at each stage of the cycle (reactor, partitioning, manufacturing), during the transition phase and at the equilibrium level. This scenario is declined according to the diagram in Figure 3.

The current nuclear park is replaced between ~2020 and ~2050 by a mixed nuclear park: of Generation III EPR<sup>TM</sup> reactors and 33% of Generation IV SFR. From 2080 to 2100, the EPR<sup>TM</sup> 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 around 2040. The minor actinides are recycled in SFR from that time [9].

In the case of heterogeneous transmutation, a 20% MA content was studied, this content allowing MA management in one or two radial blankets.

In the case of homogeneous transmutation, the maximum content is around 4% during the transient, the equilibrium content is around 1%, the classical level for a pure SFR MA recycling.

**Figure 3: Scenario of principle with fast reactor deployments**


### Reactor and fuel cycle impact

To compare the two different methods of minor actinide management (homogeneous and heterogeneous), we choose to load in the core the same minor actinide isotopic content using the two options. One family with all MA (Np, Am, Cm) and the other with only Am.

For the heterogeneous option [10] the core is loaded with traditional MOX fuel with only a small fraction of  $^{241}\text{Am}$  coming from  $^{241}\text{Pu}$  decay (see Table 1). The radial blankets are loaded using depleted  $\text{UO}_2$  with 20% minor actinide content in volume. Two different isotopic compositions were used (see table below). The residence time of the blanket is expected to be twice that of the fuel assemblies. This value is related to fuel behaviour under irradiation (swelling and fuel/clad gap closure). The number of blanket S/A in the core is 84 (14<sup>th</sup> row).

For the homogeneous option,  $\text{UPuO}_2$  S/A are replaced by  $(\text{U}+\text{Pu}+\text{MA})\text{O}_2$  fuel. The initial plutonium part remains constant while uranium is substituted by minor actinides with respect to the total mass to be loaded in the core. The initial core fuel isotopic content corresponds to the estimated available French stocks in the year 2035 (see Table 4).

**Table 4: MA isotope content (wt.%)**

	Am	MA
$^{237}\text{Np}$		16.86
$^{241}\text{Am}$	66.67	60.62
$^{242}\text{Am}$		0.24
$^{243}\text{Am}$	33.33	15.7
$^{242}\text{Cm}$		0.02
$^{243}\text{Cm}$		0.07
$^{244}\text{Cm}$		5.14
$^{245}\text{Cm}$		1.26
$^{246}\text{Cm}$		0.09

The transmutation performances are given in Table 5 for the first step of recycling and compare heterogeneous and homogeneous for all MA or only Am management.

The charged MA mass is quiet similar in both approaches, around 2.5 tonnes per reactor. The residence time is different for the two methods (respectively 2 050 and 4 100 EFPD). MA mass discharged is lower using the heterogeneous approach (~15%) due to a higher transmutation rate in the radial blanket. But when we brought it back to the service provided (i.e. the electrical output) the homogeneous option appears more efficient. Nevertheless, the transmutation performances

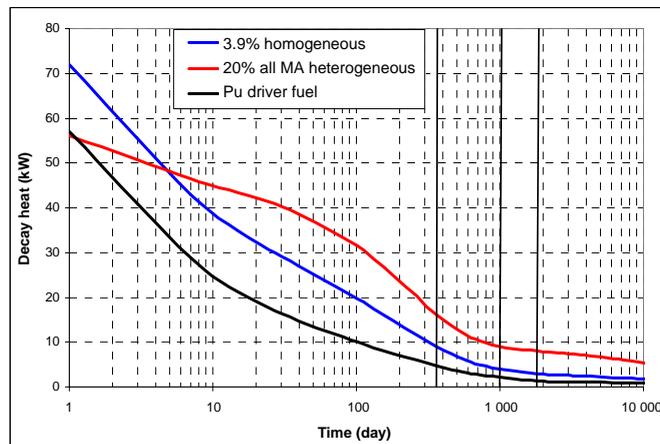
**Table 5: Transmutation performances**

Mode	Homogeneous		Heterogeneous	
Case	3.2% Am	3.9% MA	20% Am	20% MA
<b>Charged mass (kg)</b>				
Np	0	618	0	411
Am	2 429	2 258	2 438	1 866
Cm	0	67	0	160
MA	2 429	2 943	2 438	2 437
<b>Discharged mass (kg)</b>				
Np	43	335	20	265
Am	1 435	1 312	1 493	1 101
Cm	390	281	242	223
MA	1 867	1 927	1 755	1 589
<b>Transmutation rate (%) at EOL</b>				
MA	-23.1	-34.2	-28	-34.8
<b>Mass balance (kg/TWhe) at EOL</b>				
Np	+0.60	-3.97	+0.14	-1.02
Am	-13.93	-13.26	-6.62	-5.36
Cm	+5.46	2.99	+1.7	+0.44
MA	-7.87	-14.24	-4.79	-5.94

remain close in all cases, only the AM homogeneous case seems more efficient in term of mass balance. The transmutation of americium alone inevitably produces curium, but we note that even in the case of all MA transmutation there is a net production of curium.

Concerning the plutonium production in the blankets, whatever the case, the isotopic content shows a high level of <sup>238</sup>Pu and <sup>240</sup>Pu (around 50% of the total isotopic Pu content). The <sup>238</sup>Pu comes from <sup>242</sup>Cm which is continuously produced by <sup>241</sup>Am isotope under irradiation, with a very short half-life (164 days). <sup>240</sup>Pu is also produced from alpha decay of <sup>244</sup>Cm but with a larger half-life (18 years). The real impact on the reactor could come from the decay heat and the time to handling S/A (fuel for homogeneous or radial blanket for the heterogeneous). The decay heat level is driven by the curium content in the assembly. The decay heat is at least equal to the fresh fuel thermal power which is already high for back-end problematic. In addition, the <sup>241</sup>Am transmutation produces <sup>242</sup>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 behaviour.

**Figure 4: MA S/A decay heat for all MA management**



If we make the assumption that S/A can be unloaded from the reactor vessel below 40 kW (if unloaded in sodium canisters), the necessary waiting time is 30 days for heterogeneous transmutation, 10 days for homogeneous. To give an order of magnitude, the equivalent time for the standard fuel (without MA) is three days. The impact on the reactor loading /unloading planning could be investigated to determine when this time leads to a critical path. The waiting time for decreasing the decay heat below 7.5 kW (value authorising handling in gas and fuel washing) is about 400 days for homogeneous, 2 000 days for heterogeneous (200 days for standard fuel); additional in-sodium interim storage will be needed, before washing and transportation. The back-end and front-end impact on the fuel cycle plant is summarised in Tables 6 and 7 in which are presented the neutron source and decay heat in comparison with a 12% PWR MOX fuel.

**Table 6: Front-end impact, decay heat and neutron source level at manufacturing**

	Reference		Homogeneous		Heterogeneous	
	12% PWR MOX	Driver SFR fuel	3.2% Am	3.9% MA	20% Am	20% MA
Power per S/A	1	0.4	0.8	1	2	2
Power per tonnes	1	1.3	2.5	3.1	7	7
Neutron source per S/A	1	0.4	0.7	19	1.2	58
Neutron source per tonnes	1	1.3	2.2	57	4	198

**Table 7: Back-end impact, decay heat and neutron source level at reprocessing (5-year cooling time)**

	Reference		Homogeneous		Heterogeneous	
	12% PWR MOX	Driver SFR fuel	3.2% Am	3.9% MA	20% Am	20% MA
Power per S/A	1	0.4	0.8	0.9	2.4	1.8
Power per tonnes	1	1.1	2.4	2.6	8.3	6.2
Neutron source per S/A	1	0.2	0.8	0.6	3.2	2.6
Neutron source per tonnes	1	0.6	2.4	1.7	11.2	8.9

The impact of curium recycling is underlined for both strategies; the neutron source is very high in comparison to driver fuels. It is needed to establish the appropriate biological protections, shielding and robotic. This problem is more severe for the heterogeneous method. The power per S/A seems acceptable and manageable. The transmutation of Am alone seems easier to manage. We also note that the transportation of such fresh S/A with a large amount of MA (including Cm) has not yet been demonstrated.

The impact compared to a 12% PWR MOX fuel is large in absolute terms (comparison per initial heavy nuclide) and could be problematic depending on partitioning and reprocessing issues. The impact is reduced per S/A, the multiplicative factor decrease around 3 for the neutron source and also for the decay heat. For the homogeneous case the impact remains weak.

## Conclusions

The purpose of this publication is to direct attention towards the whole issue related to the transmutation of minor actinides and does purport to be a reflection of the technological reality.

Consideration of transmutation on an industrial scale remains a technological challenge for which many blocks exist.

However, both solutions presented here (homogeneous management or heterogeneous management in specific radial blanket) allow net consumption of MA with the drawback of a net Cm production (large in case of Am alone transmutation, slight in case of all MA recycling).

Due to the high levels of the thermal power, neutron source and the equivalent dose of such S/A, the core management could be affected and specific procedures and/or additional equipments may be needed for the handling of these innovative S/A, e.g. unloading in sodium canisters, interim storage in sodium.

The transmutation of Am alone provides more flexibility and seems easier to implement than all MA management.

All these impacts will be taken into account for a global evaluation in a nuclear scenario approach to determinate the pros and cons of each strategy.

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## **Current status and future plan of research and development on partitioning and transmutation technology in Japan**

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### **Abstract**

*After a first check and review on partitioning and transmutation (P&T) technology by the Japan Atomic Energy Commission (JAEC) in 2000, significant progress was made in respective research areas of partitioning, fuel fabrication, transmutation and fuel recycling in Japan. The second check and review on P&T technology was made by the JAEC in 2008-2009 to illustrate the benefit and significance of P&T, to review the current state of P&T technology in and outside Japan, and to discuss how to conduct future research and development. The final report, issued in April 2009, mentions that the significance of P&T technology can be reduced to three points: reduction of the potential hazard, mitigation of the requirement for geological repository site, and enhancement of the options in the design of the whole system of waste disposal. The current technology levels of P&T for both FBR and ADS were evaluated. Although the technology levels of some parts of the FBR cycle system are between basic research and engineering demonstration, P&T technology in general is still in the basic research stage because of the lack of experimental data on minor actinides (MA). It was, therefore, strongly recommended to accumulate experimental data for MA as a common basis for both FBR and ADS.*

## Introduction

In Japan, the Long-term Programme for Research and Development (R&D) on Nuclide Partitioning and Transmutation (P&T) Technology, called the OMEGA programme, was launched in 1988, and its first check and review (C&R) was implemented in 2000 by the Japan Atomic Energy Commission (JAEC) [1]. Based on the results of this C&R, R&D on P&T technology has been promoted for mainly two types of fuel cycle concepts: one is the homogeneous recycling of minor actinides (MA) with plutonium in commercial fast breeder reactors (FBR), and the other is the so-called double-strata fuel cycle concept whereby MA is separately and intensively transmuted by dedicated critical/subcritical reactors such as accelerator-driven systems (ADS).

Regarding the FBR option, the study to recycle MA had been conducted as a part of the “Feasibility Study on Commercialised Fast Reactor Cycle Systems” (FS) by the former Japan Nuclear Cycle Development Institute (JNC) and the succeeding Japan Atomic Energy Agency (JAEA). In 2006, FS was shifted to a larger-scale R&D programme called the “Fast Reactor Cycle Technology Development” (FaCT) Project conducted by JAEA. The first candidate of the commercial FBR in Japan is a Na-cooled MOX-fuelled one where MA is homogeneously added by up to 5% in the fuel. This reactor concept is coupled with the advanced aqueous reprocessing, where MA is fully recovered and recycled. As the second candidate, a Na-cooled metallic-fuelled reactor is also investigated mainly by the Central Research Institute of Electric Power Industry (CRIEPI), where dry reprocessing is adopted and MA is also recycled with plutonium. Heterogeneous loading of MA in commercial FBR is also studied in a basic level as an option to mitigate the influence of recycling MA in the large-scale commercial fuel cycle.

As for the double-strata option, the former Japan Atomic Energy Research Institute (JAERI) and the succeeding JAEA have continuously conducted the various fields of R&D on the aqueous separation of MA and FP for the spent fuel of the first stratum, design study and basic data acquisition for ADS, fabrication and dry reprocessing for dedicated ADS fuel, and so on.

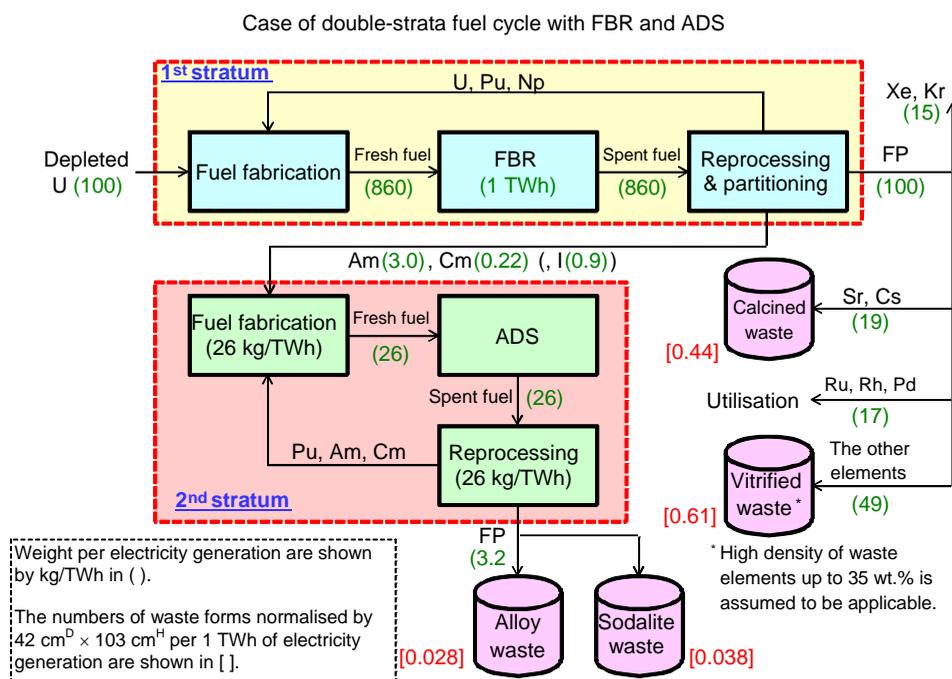
In parallel to this R&D, much effort has also been devoted to the estimation of the benefit of P&T with regard to high-level radioactive waste (HLW) management.

In August 2008, JAEC launched the Subcommittee on P&T Technology under the Advisory Committee on R&D, and conducted the second C&R to discuss the current state of the art concerning P&T technology in Japan and the future R&D programme. The Subcommittee issued its final report on 28 April 2009, and the JAEC endorsed it [2]. The final report covers the impact of P&T technology, the state of the art of the technology, an evaluation of the progress of the R&D, and recommendations for future R&D.

## Impact of P&T

In the first C&R, the urge to adopt P&T was emphasised from a viewpoint of reduction of HLW radiotoxicity. Since then, more emphasis has been placed on investigating the concrete concept of waste management incorporating P&T [3, 4], and the results were discussed in the second C&R.

Figure 1 shows an example of a fuel cycle concept and associated wastes in a case of full adoption of P&T (the case of double-strata fuel cycle with FBR in the first stratum and ADS in the second stratum). In the first stratum, uranium, plutonium and neptunium are multirecycled in the FBR, and americium and curium are partitioned to be sent to the second stratum for transmutation. At the reprocessing and partitioning plant in the first stratum, fission products (FP) are separated into three groups; Sr-Cs, platinum group metal (PGM: ruthenium, rhodium and palladium) and the other elements including lanthanides. Strontium and caesium are calcined, and stored to wait for the decrease in decay heat. It may be possible to utilise their heat and radiation rays during storage. The separated PGM may be utilised as a catalyst. Iodine is separated in a reprocessing plant, and disposed of as low-level long-lived waste or transmuted in ADS. The other waste elements from the first stratum are supposed to be vitrified with higher density (~35 wt.%) than is conventional (~15 wt.%) because of low heat generation and lack of PGM, which is undesirable for the glass melting process.

**Figure 1: An example of fuel cycle and generated wastes in case of full P&T adoption**

In the second stratum, nitride fuel (Pu, MA)N + ZrN is loaded to ADS and its spent fuel is reprocessed by pyro-chemical process, where enriched nitrogen (99%  $^{15}\text{N}$ ) will be used to restrict production of long-lived  $^{14}\text{C}$ . As the wastes from the second stratum, alloy wastes and sodalite wastes are produced. The alloy waste consists of insoluble fission products, hull and inert matrix ZrN, and therefore it is high-level waste generating low heat and can be compactly disposed of. The sodalite waste, on the other hand, is high-level waste containing heat-generating FP. Its amount is, however, relatively low, about 6% of waste from the first stratum, and therefore the wastes from the second stratum have only a limited effect on the whole system of waste management.

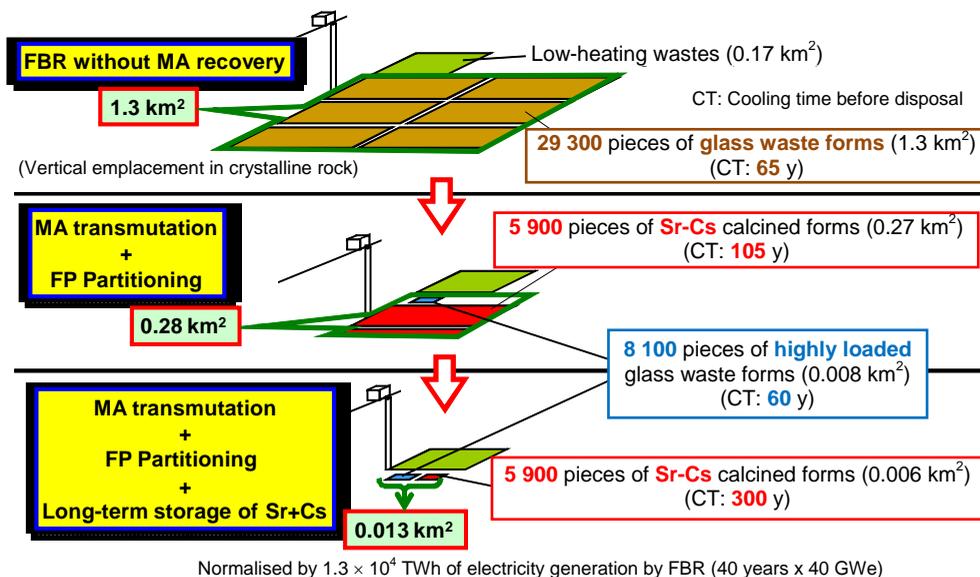
In the conventional Japanese scheme of HLW management, all the high-level liquid waste is vitrified together and cooled for 50 years before disposal. Even after the cooling period, each vitrified waste package generates heat at about 350 W, and hence is disposed of with appropriate spacing mainly in order to prevent the deterioration of confinement performance of the deep underground repository; the typical spacing is  $44 \text{ m}^2$  per one vitrified waste package in the case of vertical emplacement in crystalline host rock [5].

The conventional reprocessing for spent fuel from light water reactors (LWR) generates about 3.4 pieces of vitrified waste packages per 1 TWh of electricity generation, and therefore the area of geological repository for 1 TWh is about  $150 \text{ m}^2$ . When LWR is fully replaced by FBR, the number of the vitrified waste packages per 1 TWh is expected to be reduced by about 40% because of high energy efficiency and low yield of heat-generating FP. About 40% larger spacing is, however, necessary for the vitrified waste of FBR spent fuel because of higher accumulation of heat-generating and long-lived MA, unless they are recovered. In such a case, therefore, the effect of replacement of LWR to FBR is cancelled out. On the other hand, if MA is recovered and recycled, the necessitated spacing per one vitrified waste package will be kept at  $44 \text{ m}^2$ , and therefore the area per unit electricity generation can be reduced by about 40%.

In addition to this impact of MA transmutation, the introduction of the innovative P&T scheme such as shown in Figure 1 possibly provides a much more flexible waste management concept. The vitrified waste package without Sr-Cs or MA is no longer high heat-generating and therefore 50 years of cooling time is unnecessary; 5 years are sufficient to dispose of them by

conventional spacing ( $44 \text{ m}^2/\text{package}$ ). Even a very compact disposal concept ( $0.95 \text{ m}^2/\text{package}$ ) is applicable, supposing a cooling time of 60 years. The high heat-generating waste, Sr-Cs, is not so easy to manage. We should wait for its decay (half-life:  $\sim 30$  years) for at least 100 years to dispose of it with conventional spacing. Figure 2 compares the repository area for three cases. The upper figure is for FBR without MA recovery, where  $1.3 \text{ km}^2$  will be necessary for 29 300 packages of vitrified waste originated from 40-year operation of an FBR fleet with 40 GWe in total. If MA is recycled and Sr-Cs is separately cooled for 105 years, the total repository area will be reduced to  $0.28 \text{ km}^2$ ; about 1/4 of the non-P&T case, as shown in the middle of Figure 2. If Sr-Cs is stored and cooled for a longer period, for example 300 years, its heat generation is no longer influential to the repository structure, and hence it can be disposed of in a very compact manner [6].

**Figure 2: Examples of repository area for waste disposal concepts incorporating P&T**



It is important to consider the economic aspect when we consider the long-term storage of the waste. It is, however, notable that the calcined waste form has much better thermal property than vitrified waste, and hence it is suitable for storage with natural convection air cooling.

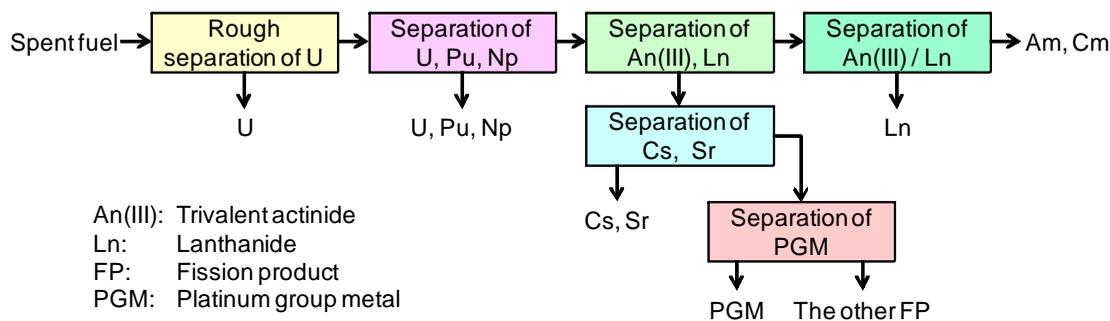
Based on these studies for various possibilities of waste management, the significance of P&T technology in the waste management system was discussed in the second C&R. The final report mentioned that three aspects should be noted:

- Introduction of P&T technology into the waste management will reduce the long-term potential radiotoxicity of the HLW. It is, however, necessary to note that this benefit is dependent upon the recovery efficiency of actinides from the waste.
- If MOX fuel is used in LWR or FBR, MA transmutation can reduce the area of repository necessitated for unit electricity generation and/or shorten the pre-disposal storage of the waste. This means that MA transmutation possibly extends the repository capacity or reduces the burden of pre-disposal storage.
- A combination of MA transmutation and FP partitioning, coupled with the long-term pre-disposal storage of the waste forms, will possibly reduce the repository area and enhance flexibility to design a more reasonable system of waste disposal by combining various options. More detailed investigation is, however, necessary to verify the feasibility of the long-term storage and the waste disposal method.

## Partitioning

R&D on partitioning process has been ongoing for spent fuel of both FBR and LWR. A conceptual flow diagram of the partitioning process is generally illustrated as in Figure 3. The first and the second processes, namely rough separation of uranium and separation of uranium, plutonium and neptunium, have been studied in the FaCT Project for the future FBR fuel cycle.

**Figure 3: Concept of partitioning process**



As for the separation process of trivalent actinide, An(III), and lanthanide from high-level liquid waste, an extraction chromatography method and a solvent extraction method have been investigated for both FBR and LWR spent fuels. For the extraction chromatography method, extractants such as CMPO (n-octyl(phenyl)-N, N-diisobutyl-carbamoyl-methylenephosphine oxide) and TODGA (N, N, N', N'-tetraoctyldiglycolamide) have been investigated. The Transuranium Extraction (TRUEX) method has also been studied and is considered as a back-up option. Moreover, for a solvent extraction method, innovative extractants have been proposed based on a "CHON principle" which is aiming at the reduction of secondary waste by using only four elements: carbon (C), hydrogen (H), oxygen (O) and nitrogen (N). As a part of this investigation, TDdGA (N, N, N', N'-tetradodecyl-diglycolamide) was invented and its excellent performance was demonstrated.

As for the separation process of An(III) from lanthanide, investigation of the conventional Solvent Extraction for Trivalent f-element Intra-group Separation in CMPO-complexant System (SETFICS) method and invention of new extractants have been implemented. One of such new extractants is PDA (N, N'-dialkyl-N, N'-diphenylpyridine-2, 6-dicarboxamide), which is effective for the extraction of An(III) in high-concentration nitric acid. An extraction chromatography method using PDA was experimentally tried by using a small-size column and simulation liquid in 5N nitric acid, and separation of americium from europium as a representative of lanthanides was successfully demonstrated. As for the separation of curium from americium, an ion extraction method is being studied.

For the separation of strontium and caesium from the other FP, a solvent extraction method, an extraction chromatography method and an ion exchange method have been investigated in parallel. Among these, a series of extraction chromatography using calix-crown R14 for caesium and crown ether for strontium as extractants is considered as a first candidate because of its viability in high-concentration nitric acid. As for the separation of PGM, an electrolytic method and an adsorption method are under investigation.

By combining these progresses on separation processes, a total partitioning system is proposed. This total system has an advantage that dilution of nitric acid concentration of the waste liquid is not necessary and all the extractants do not contain phosphorus which becomes secondary waste [7].

Based on these studies on the partitioning process, the technical level of the R&D was evaluated in the second C&R, where four levels were defined as:

- *Level 1 – Feasibility study:* The concept of the technology is created, and preliminary analysis and basic experiment are under way to verify the validity of the concept.
- *Level 2 – Basic research:* Basic experiment, measurement of basic properties of materials, design evaluation by calculation, etc., are under way to consolidate the technological basis of the concept.
- *Level 3 – Semi-engineering research:* After the verification of the fundamental feasibility, small-scale experimentation considering the final stage is under way. Verification of engineering feasibility has just started.
- *Level 4 – Engineering research:* Considering the solutions for main technical challenges, development of devices, examination of the performance and experiment in realistic conditions are under way. Realisation of the technology as a whole system is strongly intended.

For the partitioning process, the final report made the following evaluations:

- Technical challenges remain with regard to extraction chromatography for americium and curium separation, and therefore it is considered to be in Level 2. To judge its feasibility in comparison with conventional solvent extraction methods, further R&D is necessary.
- R&D activities to explore innovative extractants such as TODGA are highly appreciated, as they are expected to dramatically enhance the performance of the separation process.
- Separation of strontium and caesium remains at Level 1 because this promising process, including the resultant waste form, is still under investigation. This method is considered important due to the potential reduction of the repository area; thus, it is recommended to continue R&D along with a safe and reasonable concept for pre-disposal storage.

### Fuel for MA transmutation

R&D on fuel fabrication for MA transmutation has been ongoing for FBR and ADS. For FBR, MA-bearing (<5%) MOX fuel is the first candidate. Its fabrication method is being investigated within the FaCT Project. About 200 pieces of MOX pellets with 5% americium and about 150 pieces with 2% neptunium + 2% americium were sintered, and fuel pins were successfully fabricated to irradiate them in the experimental fast reactor JOYO. Ten-minute and 24-hour irradiations of these MA-bearing MOX fuels were carried out, and post-irradiation examinations are under way. Moreover, CRIEPI conducted the fabrication of metallic fuels such as U-Pu-Zr+2%MA+2%RE, U-Pu-Zr+5%MA and U-Pu-Zr+5%MA+5%RE at the Institute for Transuranium Elements (ITU), and they were successfully irradiated in Phénix. Higher loading of MA (~20%) into MOX fuel for heterogeneous recycling of MA in commercial FBR is also being studied.

As for dedicated transmutation fuel for ADS, study on MA nitride fuel has continued. Various compositions of MA nitride such as (Pu, Am)N, (Np, Pu, Am)N and (Np, Pu, Am, Cm)N were synthesised by a carbothermic reduction method, and their thermal properties were measured. To use these nitride fuels in ADS, inert matrix (or diluent) is added to adjust the power density. ZrN and TiN are being investigated to act as such an inert matrix, and (Pu, Am, Zr)N, (Pu, Am, Cm, Zr)N and (Pu, Am)N+TiN were synthesised and their thermal properties were measured.

Based on these studies for a MA transmutation fuel, the technical level was evaluated in the second C&R:

- *MA-bearing MOX fuel:* The fabrication method itself seems to be at Level 3. Its verification, however, has not yet been undertaken using MA including curium, and therefore the technical challenges regarding the heat generation and high radiation field caused by MA remain to be solved. Hence the technical level is considered between the Levels 2 and 3.

- *MA-bearing metallic fuel*: The fabrication method itself seems to be at Level 3. The technical challenges concerning MA remain to be overcome, and hence the technical level is considered between Levels 2 and 3.
- *Dedicated nitride fuel*: The principle of the fabrication method was verified, but investigation to extend it to the engineering scale has not been conducted. Therefore, the technical level is considered to be at Level 2.

## Transmutation system

A design study on a large-scale (~1 500 MWe) MOX FBR has been conducted as a part of the FaCT Project. Coolant void reactivity and Doppler reactivity, which are important to avoid prompt critical state in case of a hypothetical accident, can satisfy the criteria if MA content is limited below 5%. For metallic-fuelled FBR, a similar result was obtained.

As for the ADS, an 800 MWth lead-bismuth eutectic (LBE)-cooled tank-type subcritical system driven by a 30 MW superconducting proton linac with a beam energy of 1.5 GeV was proposed [8]. To realise such a large-scale ADS, various technical challenges in neutronics design, structural design, materials, beam trip transient, LBE technology and so on, should be overcome. To prevent fuel claddings made of Mod. 9Cr-1Mo steel from corrosion by LBE, the maximum surface temperature of the fuel cladding was set at 550°C with an averaged coolant inlet/outlet temperature of 300/400°C. To satisfy this criterion, a large power peaking factor caused by the subcriticality should be mitigated. It was achieved by adopting four-zone fuel loading and adjustment of inert matrix contents in the fuel, which was also beneficial to minimise alteration of subcriticality due to burn-up [9]. A beam window is subjected to pressure of LBE, radiation and high-temperature damage by protons and neutrons. A design concept of a beam window to manage these technical challenges has been investigated [10], but experimental verification especially on the irradiation damage remains to be done. Frequent beam trips will cause thermal fatigue of structural materials in the subcritical reactor and decrease plant availability down to unacceptable level. So as to determine the current technical level of the reliability of the accelerator, the beam trip frequency of an ADS plant was estimated on the basis of operational information of existing accelerators, and was compared with acceptable trip frequency of subcritical reactors as a function of the beam trip duration ( $\tau_{\text{trip}}$ ). It was shown that frequency of short beam trips ( $\tau_{\text{trip}} < 10$  s) is already within the limitation, and medium ( $10 \text{ s} < \tau_{\text{trip}} < 5$  min.) and long ( $5 \text{ min.} < \tau_{\text{trip}}$ ) ones should be decreased by a factor of 7 and 30, respectively [11]. This reduction is expected to be possible by shortening the beam trip duration, enhancing reliability of accelerator components, taking preventive measures automatically based on in-service diagnosis, and mitigating criteria by subcritical reactor design. Experimental study is also continued on corrosion behaviour of material by using an LBE test loop and a static corrosion test device, velocity profiling of LBE by an ultrasonic Doppler method, properties of irradiated materials, and so on.

Based on these studies for the transmutation systems, the technical level was evaluated in the second C&R:

- *Transmutation by FBR*: The current accuracy of MA nuclear data is satisfactory for conceptual study but not sufficient for evaluation of safety parameters. It is recommended to conduct integral experiments using 10 kg order of americium and curium, which is an important issue for both FBR and ADS.
- *Transmutation by ADS*: Remarkable progress was seen, but accumulation of basic data is still necessary to proceed to the engineering verification. The technical level is, therefore, considered to be at Level 2.

## Transmutation fuel recycling

The aqueous process for MOX fuel of FBR is being developed under the FaCT Project and was briefly discussed in the previous section. Dry processes for metallic fuel of FBR and nitride fuel of ADS are also being studied mainly by CRIEPI and JAEA, respectively. As for metallic fuel, a small-scale electrolytic refining test for unirradiated U-Pu-Zr metallic fuel was carried out, and dissolution of fuel on an anode and deposition behaviour of uranium and plutonium on a cathode were verified.

As for nitride fuel, electrolytic refining tests for AmN, UN with elements simulating fission products, and UN and PuN with inert matrix diluents, and so on, were carried out. Moreover, a series of experiments to recover uranium and plutonium in a cadmium cathode, to nitride them and to fabricate (U, Pu)N pellets were successfully carried out.

Based on these studies for transmutation fuel recycling, the technological level was evaluated in the second C&R:

- *Dry process of metallic fuel for FBR:* The technical level is in Level 3 as a whole, though a part of the technology is already at Level 4. It is recommended to accumulate further basic data for peripheral technologies such as the process of salt waste.
- *Dry process of nitride fuel for ADS:* The activities are still at the stage of principle verification at laboratory scale, and therefore the technical level is at Level 2. It should be noted that since this technology has many common aspects with metallic fuel, a collaborative approach for these two fuel types is recommended.

## Recommendations for future R&D

The final report of the second C&R stated recommendations for future R&D on P&T technology in Japan:

- R&D activities on P&T should be linked strongly with those of FBR because the purpose of P&T is identical to one of the targets that FBR is expected to achieve in the future. From this viewpoint, R&D should be promoted not to explore the maximum effect of P&T but to achieve the required total performance of the future energy system on safety, economy, environmental sustainability, resource saving and proliferation resistance. This is also true for the double-strata fuel cycle concept; the merit of isolating the dedicated transmutation cycle from the commercial power generation cycle should be carefully discussed.
- Although the technology is growing from the basic research level to a semi-engineering level in general, information to evaluate the performance of the whole system is not sufficient. Fundamental data, therefore, should be accumulated and expanded.
- Two types of concepts, the homogeneous FBR fuel cycle and the double-strata fuel cycle, should be promoted in a strongly co-ordinated manner. Results of periodical assessment of the system performance should be reflected in their R&D activities.
- For P&T technology using the homogeneous MOX FBR fuel cycle, future important issues are reliable MA separation process, feasible fuel fabrication process under high heat generation and high dose rate, safety design of the reactor core containing MA up to 5%, and reliable fuel performance for high burn-up. For metallic-fuelled FBR, R&D should be continued to verify its engineering feasibility.
- For the double-strata fuel cycle concept, future important issues are an accelerator with sufficient safety, reliability and economy, feasibility of a beam window, reactor physics of a subcritical core including its controllability, design of an LBE-cooled subcritical reactor, feasibility of the dry process for nitride fuel, and reliable fabrication of nitride fuel with appropriate performance.

- For separation processes in common, it is important to accumulate knowledge and experience through hot experiments at laboratory scale, cold examination of devices at engineering scale and demonstrative testing using simulated high-level liquid waste, before proceeding to hot demonstration of the technology at engineering scale.
- For MA-bearing fuels in common, a database of material properties of fuels including curium is not sufficient, and it is not possible to judge the engineering feasibility to fabricate it under high heat generation and high dose rate. It is, therefore, important to continue the acquisition of fundamental data.
- For transmutation systems in common, fundamental data to evaluate safety and performance of the systems are not sufficient at present; it is therefore recommended to enhance activities on MA nuclear data in existing facilities and to provide an experimental facility to conduct reactor physics experiments using MA. The Transmutation Physics Experimental Facility proposed under the J-PARC Project is one of the candidates to conduct such an experiment. For ADS, further accumulation of fundamental data is necessary by collaborating upon the same kinds of activities overseas, development of accelerator neutron sources and development of FBR. J-PARC is expected to play an important role in conducting an accelerator-reactor coupling experiment.
- To continue R&D on P&T technology, it is important to provide assessment tools for the performance of the whole system, to facilitate co-ordination between basic research and engineering development (vertical collaboration) and to facilitate interdisciplinary synergy among different areas of science and engineering (horizontal collaboration).
- C&R on the FaCT Project is scheduled for 2010. This seems a good opportunity to define a concrete R&D plan for P&T technology. It is recommended to propose reasonable and strategic plans for acquisition of fundamental data, benchmark experiments and provision of experimental facilities.

## Conclusion

R&D on P&T technology showed significant progress in Japan in the research areas of partitioning, fuel fabrication, transmutation and fuel recycling. The second C&R was conducted by JAEC in 2008-2009 to illustrate the benefit and significance of P&T, to review the current state of P&T technology, and to discuss how to conduct future research and development.

The final report mentions that the significance of P&T technology can be reduced to three points: reduction of the potential hazard, mitigation of the requirement for geological repository site, and enhancement of the options in the design of the whole system of waste disposal.

The current level of P&T technology was evaluated. Although the technical levels of some parts of the FBR cycle system are between basic research and engineering demonstration, P&T technology in general is still at the basic research level because of the lack of experimental data for MA. It was, therefore, strongly recommended to accumulate experimental data for MA as a common basis for both FBR and ADS.

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## **Session II**

### **Waste Forms and Geological Disposal**

***Chairs: W.M. Nutt, I-S. Hwang***



## Fuel cycle potential waste for disposition

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### Abstract

*The United States currently utilise a once-through fuel cycle where used nuclear fuel (UNF) is stored on-site in either wet pools or in dry storage systems with ultimate disposal in a deep mined geologic repository envisioned. Within the Department of Energy's (DOE) Office of Nuclear Energy (DOE-NE), the Fuel Cycle Research and Development Program (FCR&D) develops options for the current commercial fuel cycle management strategy to enable the safe, secure, economic and sustainable expansion of nuclear energy while minimising proliferation risks by conducting research and development of advanced fuel cycles, including modified open and closed cycles.*

*The safe management and disposition of used nuclear fuel and/or nuclear waste is a fundamental aspect of any nuclear fuel cycle. Yet, the routine disposal of used nuclear fuel and radioactive waste remains problematic. Advanced fuel cycles will generate different quantities and forms of waste than the current LWR fleet.*

*This study analyses the quantities and characteristics of potential waste forms, including differing waste matrices, as a function of a variety of potential fuel cycle alternatives including:*

- *Commercial UNF generated by uranium fuel light water reactors (LWR). Four once-through fuel cycles analysed in this study differ by varying the assumed expansion/contraction of nuclear power in the United States.*
- *Four alternative LWR used fuel recycling processes analysed differ in the reprocessing method (aqueous vs. electrochemical), complexity [Pu only or full transuranic (TRU) recovery] and waste forms generated.*
- *Used mixed-oxide (MOX) fuel derived from the recovered Pu utilising a single reactor pass.*
- *Potential waste forms generated by the reprocessing of fuels derived from recovered TRU utilising multiple reactor passes.*

## Introduction

The safe management and disposition of used nuclear fuel and/or nuclear waste is a fundamental aspect of any nuclear fuel cycle; however, the routine disposal of used nuclear fuel and radioactive waste remains problematic. Advanced fuel cycles will generate different quantities and forms of waste than the current light water reactor (LWR) fleet. This study analyses the quantities and characteristics of potential waste forms including differing waste matrices, as a function of a variety of potential fuel cycles. The information presented in this paper is more completely investigated in Refs. [1, 2].

### Commercial light water reactor used nuclear fuel – once-through fuel cycle

Commercial nuclear power plants have operated in the United States since about 1960. There are currently 104 operating nuclear power plants. Used nuclear fuel (UNF) from these operating plants is currently stored on-site in pools or dry storage casks with disposal in a geologic repository envisioned in a once-through fuel cycle. In addition, UNF from 14 shutdown reactors is currently stored on the reactor sites and at the General Electric wet storage facility at Morris, IL.

### Current LWR uranium oxide used fuel inventory

The source of current inventory data for this study is information collected in support of the Department of Energy Office of Civilian Radioactive Waste Management's (OCRWM) efforts for licensing the Yucca Mountain Repository [3]. Information collected from RW-859 forms is available on an assembly basis for UNF discharges from 1968 through 2002. Data is also available that was collected to support RW activities on a batch basis for fuel discharges from 1968 through April 2005 [4].

To develop an inventory estimate through 2009, fuel discharge predictions developed for the Nuclear Energy Institute in 2005 were used to estimate the number of assemblies and metric tonnes of uranium [5]. To estimate the average enrichment and burn-up through 2009, projections made by utilities as part of the RW-859 surveys were used. These projections are documented in OCRWM's "Calculation Method for the Projection of Future Spent Fuel Discharges" [6]. These projections identified a burn-up increase of 2.38% per year for boiling water reactor (BWR) fuel and 1.11% per year for pressurised water reactor (PWR) fuel through 2009. The enrichment increased at the same rate as burn-up. Comparison of these projections made in 1998 to actual data collected through 2004 show very good agreement (PWR – actual 46 950 MWd/MTU vs. projected 46 922 MWd/MTU; BWR – actual 43 447 MWd/MTU vs. 42 787 projected MWd/MTU). Table 1 provides an estimate of the commercial UNF discharged through December 2009. Figure 1 provides a distribution of this estimated inventory as a function of burn-up. Nearly 100% of the fuel currently being discharged exceeds the "high burn-up" threshold of 45 000 MWd/MT.

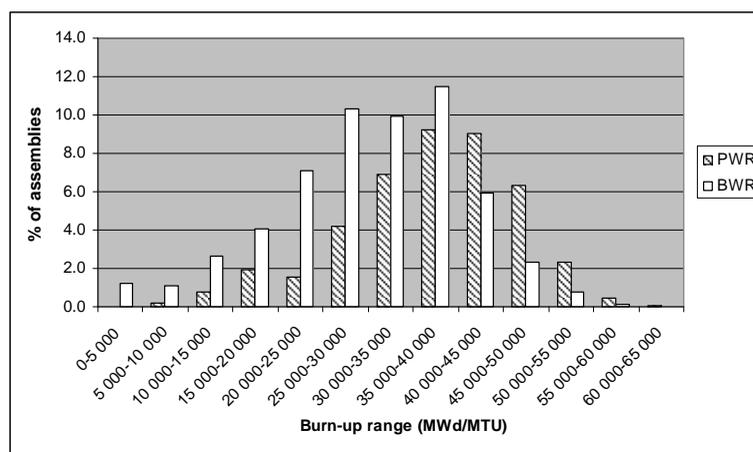
**Table 1: Commercial UNF estimated discharge through 2009**

Total No. of assemblies <sup>a</sup>			Total initial uranium (MTU) <sup>b</sup>			Average enrichment		Average burn-up (MWd/MTU) <sup>c</sup>	
PWR	BWR	Totals	PWR	BWR	Totals	PWR	BWR	PWR	BWR
94 800	124 000	218 800	41 100	22 100	63 200	3.72	3.07	39 400	32 700

<sup>a</sup> The number of assemblies has been rounded to the nearest 200, totals may not appear to sum correctly.

<sup>b</sup> The estimated fuel discharged has been rounded to the nearest 100 metric tonnes of uranium (MTU), totals may not appear to sum correctly.

<sup>c</sup> The burn-up has been rounded to the next 100 megawatt-days/MTU (MWd/MTU).

**Figure 1: Percentage of assemblies per burn-up range in the current inventory**

### Future LWR uranium oxide used fuel projections

Future UNF discharge predictions were developed using the same methods used for estimating the current UNF inventory. These projections identified a burn-up increase of 2.38% per year for BWR UNF and 1.11% per year for PWR UNF. The enrichment increased at the same rate as burn-up until reaching the current enrichment limit of 5%. Once the 5% enrichment limit is reached, the enrichment and burn-up are assumed to remain constant.

To provide a range of UNF requiring disposal, four nuclear energy scenarios were selected from those previously evaluated by DOE:

- *Scenario 1* assumes no replacement of existing nuclear generation reactors. The existing plants are assumed to have one 20-year life extension and will be decommissioned after 60 years of operation. Applying these assumptions the last nuclear generator finishes operations in 2055.
- *Scenario 2* assumes the amount of current nuclear generation is maintained at the current levels [100 gigawatt-electric/year (GWe/yr)] with new reactors replacing the existing reactors as the existing reactors are decommissioned. The current PWR/BWR ratio is maintained.
- *Scenario 3* assumes the amount of nuclear generation will increase to 200 GWe/yr from 2020 to 2060.
- *Scenario 4* assumes the amount of nuclear generation will increase to 400 GWe/yr from 2020 to 2060.

In both Scenarios 3 and 4 new nuclear generating capacity is assumed to come on-line in 2020 and increase linearly until 2060. Nuclear generation is assumed to remain constant past 2060. Table 2 provides the mass (metric tonnes of uranium) projected to be generated annually in 2060 (at the assumed completion of nuclear expansion) and cumulatively through the end of the century.

**Table 2: Commercial UNF projections**

	Scenario			
	No replacement	100 GWe/yr	200 GWe/yr	400 GWe/yr
Annual generation rate at 2060	0	2 300	4 500	9 100
Cumulative UNF (MTU)	140 000	270 000	407 000	682 000

## Process waste generated by reprocessing commercial light water reactor fuel

To provide a tool for evaluation of the impact of reprocessing, unit quantities of 24 representative LWR UNF compositions have been converted into their equivalent waste forms for a variety of reprocessing methods. For each of the representative fuels the mass, volume, container count and decay heat in each container have been projected for each of the baseline waste forms anticipated.

### Reprocessing methods

Commercial light water reactor UNF reprocessing methods vary in process complexity and technical maturity. Generally the objective of additional complexity is to lessen the potential environmental impact of the resulting waste disposition activities. To support future evaluations of potential environmental impact three aqueous reprocessing methods and one electrochemical reprocessing method were selected:

- *Co-extraction* represents the simplest and most technically mature aqueous reprocessing method evaluated. The process envisioned is similar to the current generation of deployed reprocessing technology (e.g. the Rokkasaho Reprocessing Facility). Uranium and plutonium are recovered together (no pure plutonium separation). The principle fission product wastes including the minor actinides are combined with the undissolved solids (UDS) and recovered Tc into a single borosilicate glass waste form. The gaseous radionuclides  $^{129}\text{I}$  and  $^3\text{H}$  released during reprocessing are captured and converted to waste forms suitable for disposal while  $^{14}\text{C}$  and  $^{85}\text{Kr}$  are assumed to be released to the atmosphere.
- *New extraction* is an advanced aqueous process which recovers all of the transuranic (TRU) elements for re-use. The process envisioned includes transuranic extraction (TRUEX) and the trivalent actinide lanthanide separation by phosphorus-based aqueous complexes [sic.] (TALSPEAK) process for complete TRU recovery. The principle fission product wastes are combined with the UDS and separated Tc into a single borosilicate glass waste form. The principle gaseous radionuclides  $^{129}\text{I}$ ,  $^{85}\text{Kr}$ ,  $^{14}\text{C}$  and  $^3\text{H}$  released during reprocessing are captured and converted to waste forms suitable for disposal.
- *Uranium extraction (UREX)* is an advanced aqueous process which also recovers all of the TRU and in addition separates the fission product waste components into three segments. The fission product extraction (FPEX) process is added to separate the Cs/Sr/Ba/Rb, which is converted to a solid ceramic waste form. The Tc and undissolved solids (UDS) are combined with a portion of the zirconium hulls/stainless steel hardware to form a metal alloy, and the remaining fission products are converted to a borosilicate glass. UREX is the most complex of the three aqueous processes evaluated.
- *Electrochemical processing* is a dry process using conductive molten salt baths to recover all the TRU elements. In this process the fission products are split between three waste streams. Elements which are more noble (as measured by electrochemical potential) than uranium, such as fuel cladding and noble metal fission products, remain as metals and are incorporated into a metal alloy waste form. Elements less noble than uranium are converted to chloride salts. The lanthanide elements are recovered from the salt by electrolysis and converted to a lanthanide glass. Excess salt is purged; the chloride is adsorbed by zeolite and bonded with glass to make the final waste form. The principle gaseous radionuclides  $^{129}\text{I}$ ,  $^{85}\text{Kr}$ ,  $^{14}\text{C}$  and  $^3\text{H}$  released during reprocessing are captured and converted to a waste form suitable for disposal, although most of the  $^{129}\text{I}$  in this process is not released to the gaseous phase but is converted to a molten salt and purged with the excess salt.

### **Representative fuels**

The historical and projected UNF inventories were reviewed to select fuels representative of the anticipated fuel type, burn-up and age at the time of reprocessing. Three fuel burn-ups were selected to represent the historical and future PWR (20, 40 and 60 GWd/MTU) and BWR (15, 30 and 50 GWd/MTU) reactors discharge. The time of reprocessing is unknown and a broad range of potential ages (5, 30, 100 and 500 years) were selected. This broad range of potential burn-ups and ages allows evaluation of various strategic decisions.

### **Baseline and alternative waste forms**

The Global Nuclear Energy Partnership Integrated Waste Management Strategy Baseline Study [7] summarised the state of the art in stabilisation concepts for by-product and waste streams, and recommended a baseline of waste forms for the safe disposition of proposed waste streams from future fuel recycling processes. This baseline has been adopted for this study as applicable to the specific reprocessing method.

The baseline waste forms include four specific to the principle radionuclides of interest ( $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{85}\text{Kr}$  and  $^{129}\text{I}$ ) released during fuel chopping and other front-end processing. There is considerable uncertainty regarding the need to capture and treat both the  $^{14}\text{C}$  and  $^{85}\text{Kr}$  released during reprocessing, with many factors influencing the final decision (e.g. reprocessing facility location and environmental regulations). These waste forms provide a comprehensive range of the waste quantities for potential disposal.

The baseline includes two metal waste forms for separated structural components of the fuel assemblies. These include simple compaction (following decontamination) and a more advanced metal alloy which has been proposed for the UREX process that contains the UDS and Tc. A portion of the hulls and hardware are diverted from the compaction line to provide the zirconium and iron required to produce a durable waste form.

A metal alloy is also used in the electrochemical process. Those elements which are more noble (as measured by electrochemical potential) than uranium such as the hulls, hardware and noble metal fission products remain as metals. The metal waste is decontaminated by volatilising adhered salts.

Four fission product waste forms are included. Borosilicate glass is the accepted standard for reprocessing waste disposal; however, the waste loading is limited to avoid the formation of multi-phase glasses. These limits include: i) a maximum decay heat of 14 000 watts per 2 ft diameter canister to prevent the canister centreline temperature from reaching the transition temperature; ii) the molybdenum trioxide solubility is limited to 2.5% by weight; iii) the noble (Ag, Pd, Rh, Ru) metals are limited to 3% by weight. The limit selected for any representative fuel allows the maximum waste loading and minimum projected waste volume and mass.

UREX processing proposed separating the Cs/Sr/Ba/Rb to segregate the high-heat-producing waste. The waste is treated by mixing the waste with bentonite clay followed by high-pressure pressing and high-temperature sintering to produce a ceramic puck.

The electrochemical process utilises two fission product waste forms. Excess salt is purged with fission products which have been adsorbed onto zeolite. Additional zeolite is added to sequester the excess salt chloride and then bonded with borosilicate glass. The electrochemical process also separates lanthanides which are converted to a lanthanide-based glass.

The complete results of this analysis are provided in Ref. [1]. Table 3 summarises the borosilicate glass fission product waste form generated by the co-extraction and new extraction reprocessing methods. Trends include:

- Projections of borosilicate glass quantity from the co-extraction process are limited by decay heat (14 000 watts per canister) when “young” UNF is processed.
- Projections of borosilicate glass quantity from the co-extraction processing of UNF older than 30 years are limited by molybdenum trioxide solubility at 2.5 wt.%. In these cases

the mass, volume and containers per metric tonne are constant regardless of the age of the UNF processed, although the decay heat continues to decline with fuel age.

- Many of the fission product waste forms included in this study significantly exceed the 1 500 watts/canister limit of the Yucca Mountain license application. Disposal in another undetermined alternative repository could require additional decay storage time prior to disposal or a more dilute waste form.
- Recovery of the Am/Cm in other aqueous reprocessing methods and separation of the Cs/Sr in the UREX reprocessing (results provided in Ref. [1]) reduces the decay heat such that the waste loading is limited by molybdenum trioxide solubility at 2.5 wt.%. In these cases the mass, volume and containers per metric ton are constant regardless of the age of the UNF processed, although the decay heat continues to decline with fuel age.

Trends in the captured off gas and metal waste forms are discussed in Ref. [1] along with nine alternative waste forms.

**Table 3: Fission product waste trends for pressurised water reactor used nuclear fuel for co-extraction and new extraction recycling methods**

		Co-extraction				New extraction			
		Borosilicate glass				Borosilicate glass			
		Containers: 2 ft diameter × 15 ft tall canisters. Each canister contains 2 900 kg.				Containers: 2 ft diameter × 15 ft tall canisters. Each canister contains 2 900 kg.			
Burn-up (GWd/MTU)	Age (years)	Mass (kg/MT)	Volume (ft <sup>3</sup> /MT)	Containers per MT	Decay heat (W/container)	Mass (kg/MT)	Volume (ft <sup>3</sup> /MT)	Containers per MT	Decay heat (W/container)
20	5	198.53	3.22	0.07	14 000	139.30	2.26	0.05	12 124
	30	147.61	2.40	0.05	7 766	139.30	2.26	0.05	2 911
	100	147.61	2.40	0.05	3 059	139.30	2.26	0.05	562
	500	147.61	2.40	0.05	1 004	139.30	2.26	0.05	0
40	5	410.33	6.66	0.14	14 000	253.53	4.12	0.09	13 111
	30	268.66	4.36	0.09	8 367	253.53	4.12	0.09	3 157
	100	268.66	4.36	0.09	2 928	253.53	4.12	0.09	601
	500	268.66	4.36	0.09	884	253.53	4.12	0.09	0
60	5	658.47	10.69	0.23	14 000	366.12	5.95	0.13	13 851
	30	387.97	6.30	0.13	8 667	366.12	5.95	0.13	3 233
	100	387.97	6.30	0.13	2 546	366.12	5.95	0.13	611
	500	387.98	6.30	0.13	654	366.13	5.95	0.13	0

### **Mixed-oxide used nuclear fuels**

Mixed-oxide (MOX) fuel is a blend of oxides of plutonium and natural uranium, reprocessed uranium or depleted uranium. This study examines two potential sources of fissile material.

Recycling LWR uranium oxide (UOX) used fuel (burnt to 51 GWd/MT and allowed to cool for five years post-irradiation) recovers plutonium and uranium which can then be fabricated into a MOX fuel. This MOX fuel is stored for two years prior to introduction into the full MOX core. The delay time results in the build-up of <sup>241</sup>Am in the MOX fuel, which arises from the decay of <sup>241</sup>Pu. The burn-up of the MOX core is limited to 50 GWd/MT because of a constraint on the plutonium content in the MOX fuel. The average plutonium enrichment is 10.74%; therefore, each metric tonne of LWR fuel which is reprocessed allows fabrication of 108.9 kg of MOX fuel.

Recognising the threat of surplus weapons-grade plutonium to global security, the governments of the United States of America and the Russian Federation agreed to pursue a long-term disposition initiative limiting the availability of this material for weapons use. The US MOX fuel programme will dispose of 34 metric tonnes (MT) of weapons-grade plutonium. To dispose of this quantity of plutonium the programme will produce approximately 1 684 fuel assemblies [8]. Each assembly will contain 462.2 kg of heavy metal (HM) per un-irradiated fuel assembly [8]. A total of approximately 77.8 MTHM of MOX fuel will be generated.

**Potential waste derived from reprocessing advanced burner reactor UNF**

A key attribute of the “fully closed” nuclear fuel cycle is that no UNF is disposed, only UNF reprocessing wastes are disposed. To investigate reducing the long-term (vs. transitional) TRU disposal burden on the repository, this study selected sodium-cooled fast reactors with design features and operating parameters such that a burning TRU conversion ratio of 0.5 and 0.75 are achieved. Both oxide and metal fuel forms are used and these fuel types are “associated” with aqueous and electrochemical reprocessing technologies respectively. While this association is not technically mandated, using this association does allow the differences in the reprocessing methods to be examined.

Tables 4 and 5 summarise the fission product waste forms which may be potentially generated by recycling advanced burner reactor (ABR) fuel. Off gas and metal waste forms are investigated more completely in Ref. [1].

**Table 4: Fission product waste forms from aqueous recycling advanced burner reactor UNF**

		Aqueous			
		Borosilicate glass			
		Containers: 2 ft diameter × 15 ft tall canisters. Each canister contains 2 900 kg.			
Burn-up (GWd/MTU)	CR	Mass (kg/MT)	Volume (ft <sup>3</sup> /MT)	Containers per MT	Decay heat (W/container)
<b>Oxide-based fuel</b>					
131	0.75	1 690	27.45	0.58	14 000
166	0.50	2 090	33.95	0.72	14 000

**Table 5: Fission product waste forms from electrochemical recycling advanced burner reactor UNF**

		Electrochemical							
		Glass-bonded zeolite				Lanthanide glass			
		Containers: 2 ft diameter × 15 ft tall canisters. Each canister contains 2 900 kg.				Containers: 6 in diameter × 60 in tall canisters. Each canister contains 500 kg.			
Burn-up (GWd/MTU)	CR	Mass (kg/MT)	Volume (ft <sup>3</sup> /MT)	Containers per MT	Decay heat (W/container)	Mass (kg/MT)	Volume (ft <sup>3</sup> /MT)	Containers per MT	Decay heat (W/container)
<b>Metal-based fuel</b>									
99.6	0.75	2 641	42.77	0.91	2 900	58.39	0.46	0.12	21 175
132	0.50	3 368	54.53	1.16	3 368	73.14	0.57	0.15	21 574

**Secondary waste**

Secondary waste streams will be generated from the disposition of used fuel regardless of the fuel cycle utilised. Secondary waste streams are non-process waste streams typically consisting of contaminated items such as protective clothing, other job control waste, maintenance waste and failed equipment. Secondary waste estimates have been prepared for the following used fuel disposition activities:

- geologic disposal of LWR used fuel;
- aqueous and electrochemical recycling of LWR used fuel;
- aqueous and electrochemical recycling of sodium fast reactor (SFR) used fuel.

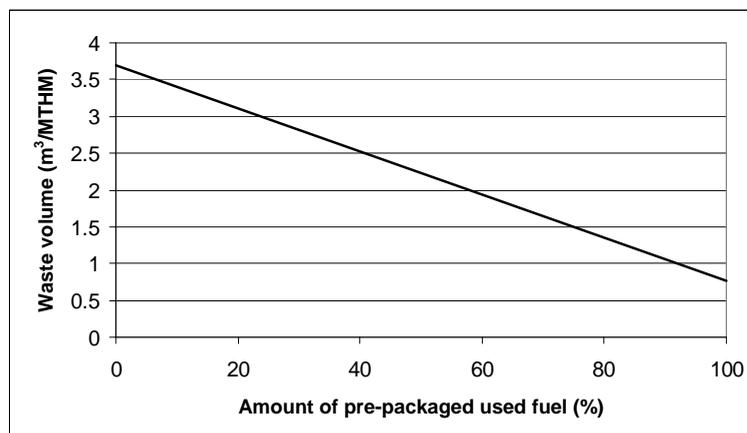
Estimates of Class A/B/C low-level waste (hereinafter referred to as LLW), greater than Class C low-level waste (GTCC waste), mixed Class A/B/C low-level waste (hereinafter referred to as mixed LLW) and mixed greater than Class C low-level waste (mixed GTCC waste) are provided.

### **Secondary waste from geologic disposal of LWR used fuel**

Secondary waste estimates were prepared as part of the Environmental Impact Statement for a geologic repository at Yucca Mountain [9]. The waste estimates prepared for the Yucca Mountain repository form the basis for the secondary waste estimates reported here.

Repository operations are expected to generate only LLW. GTCC waste, mixed LLW and mixed GTCC waste are not expected to be generated by repository operations. Estimates of LLW generation for operations at a geologic repository range from 0.8 m<sup>3</sup> per metric tonne of heavy metal (m<sup>3</sup>/MTHM) of used fuel disposed to 3.7 m<sup>3</sup>/MTHM depending on the fraction of used fuel that is pre-packaged in canisters that are suitable for direct disposal at the repository. Figure 2 shows the waste generation rate with respect to the fraction of used fuel that is pre-packaged in directly disposable canisters.

**Figure 2: LLW generated by disposal of used fuel at a geologic repository**



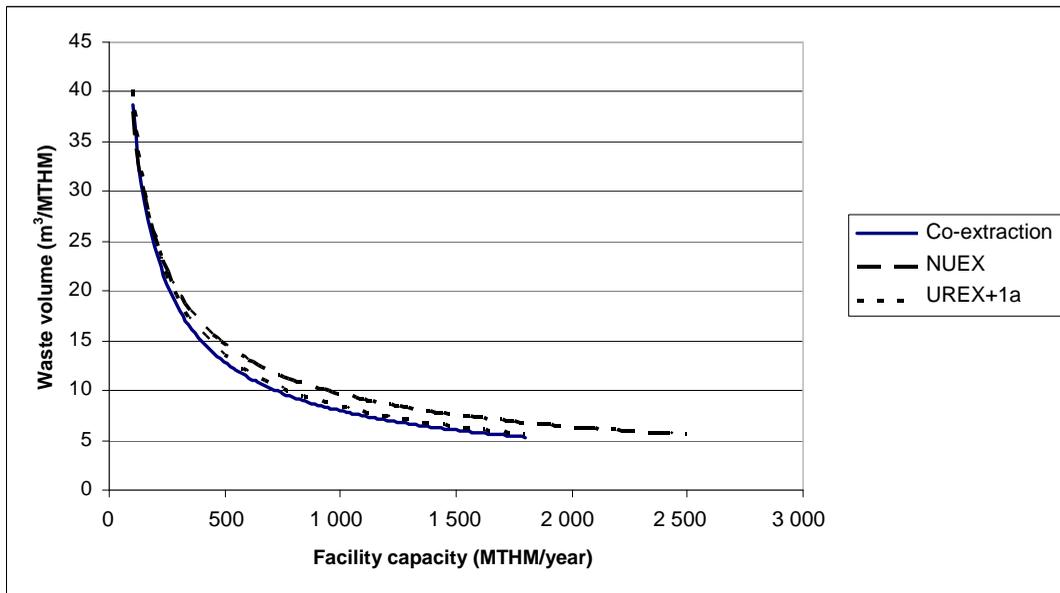
### **Secondary waste from recycling of LWR used fuel**

Secondary waste from a variety of aqueous recycling operations has been estimated relative to facility capacity. Data related to the co-extraction process proposed by AREVA [10], the new extraction (NUEX) process proposed by EnergySolutions [11] and the UREX+1a process as evaluated by the Engineering Alternative Studies (EAS) [12] were used as a basis for the estimates. Curves were generated based on the available data to provide consistent estimates for subsequent analysis of future fuel cycles. An example curve for LLW is shown in Figure 3. Similar curves for other waste types are provided in Ref. [2].

Secondary waste from electrochemical recycling of LWR used fuel has also been estimated. Table 6 shows the estimated waste volume expected from a 300 MTHM/year recycling facility based on data developed for the Engineering Alternative Studies. Secondary wastes from electrochemical recycling of LWR used fuel are investigated more completely in Ref. [2].

### **Secondary waste from recycling of SFR used fuel**

Secondary wastes from recycling of SFR used fuel does not differ substantially from the estimates for LWR used fuel. The primary differences in the estimates relate to the quantity of assemblies recycled driven primarily by the physical configuration and the radionuclide content of the fuel assemblies. Secondary wastes from recycling SFR used fuel are investigated more completely in Ref. [2].

**Figure 3: LLW generated by aqueous recycling of LWR used fuel****Table 6: LLW generated by electrochemical recycling of LWR used fuel**

Facility capacity (MTHM/yr)	Volume	LLW	GTCC	Mixed LLW	Mixed GTCC
300	m <sup>3</sup> /year	2 616.1	919	29	43.6
	m <sup>3</sup> /MTHM	8.7	3.1	0.1	0.15

### Future activities

Efforts will continue in 2011 to evaluate other fuel cycle activities including secondary wastes from the fabrication of mixed-oxide fuel. Work is also planned in conjunction with industrial partners to refine the process and secondary waste estimates already prepared for aqueous recycling of used fuel.

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## **Applying insights from repository safety assessments to evaluating impacts of partitioning and transmutation**

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### **Abstract**

*Published results of safety assessments for deep geologic disposal of high-level radioactive waste and spent nuclear fuel in the United States, Sweden, France, Switzerland and other nations provide insight into those aspects of the waste form that are most important to the long-term performance of the repository system. Hypothetical modifications to the wastes, such as might result from new technologies for processing spent fuel and advances in the design of nuclear reactors and waste forms have the potential to impact long-term performance. This paper reviews relevant results of existing safety assessments for a range of disposal concepts and provides observations about how hypothetical modifications to the waste (e.g. changes in radionuclide inventory, thermal loading and durability of waste forms) might impact results of safety assessment models. Disposal concepts considered include geologic repositories in both saturated and unsaturated environments.*

## Introduction

Deep geological repositories have been proposed worldwide for the permanent disposal of long-lived high-level radioactive waste since the middle of the twentieth century, and mined repositories are in operation for some categories of transuranic and intermediate-level waste [1, 2]. No repositories are in operation anywhere, however, for spent nuclear fuel or the high-level radioactive wastes derived from the reprocessing of spent fuel. Many factors have contributed to difficulties in siting and licensing disposal facilities, including social and political opposition from potential host communities and technical concerns focusing in part on the complexity of modelling interactions between the waste and the geologic environment and the very long time periods for which the waste must be isolated from the biosphere. Half-lives of some radionuclides are on the order of millions of years, waste can generate large amounts of heat from radioactive decay, the environment in which degradation of the waste form and its containers may occur is uncertain and estimates of the transport of radionuclides to the biosphere require characterisation of the surrounding geology.

Modifications to the waste characteristics have the potential to improve repository performance, increase the density with which waste can be emplaced in a mined repository and increase confidence in the overall safety of disposal. For example, removing heat-generating radionuclides from the waste stream and managing them separately could result in lower temperatures in the repository, allowing emplacement of significantly more reactor waste in the same mined volume [3]. Stabilising the waste in engineered materials (e.g. glasses, ceramics or metals) that degrade extremely slowly in geologic environments can increase confidence in repository performance [4] and has the potential to lower the rate at which radionuclides leave the repository. Removing transuranic actinides (primarily isotopes of plutonium, but also less abundant actinides such as neptunium, americium and curium) from the waste stream by separation and transmutation processes has the potential to significantly lower the radioactivity and radiotoxicity (defined as activity times a dose conversion factor for each radionuclide) of the waste initially emplaced in a repository. Removal of actinides from the waste form may therefore contribute to increased public confidence in the safety of geologic disposal [5], independent from its impact on estimates of potential doses to future humans.

Four safety assessments are examined in this paper as representative examples of a range of disposal concepts. The United States Department of Energy (US DOE) assessment of the Yucca Mountain site [6] evaluates disposal of both spent nuclear fuel and high-level radioactive waste in an unsaturated fractured volcanic tuff characterised by oxidising conditions and isolated seeps of water in a humid environment. Both the French *Dossier 2005* [7] and the Swiss *Project Opalinus Clay Safety Report* [8] evaluate disposal of spent nuclear fuel and high-level radioactive waste in a clay repository where conditions are anticipated to be water-saturated and chemically reducing, and are representative of approaches also being evaluated in Belgium and other countries. The Swedish *SR-Can 2006* report [9] evaluates disposal of spent nuclear fuel in reducing conditions in saturated granite, and is representative of approaches being evaluated in Finland, China, South Korea and other countries. In addition to these four national programme reports, the collaborative *RED-IMPACT* report [10] prepared in 2008 for the European Commission with contributions from 23 European partner organisations provides valuable quantitative insights into the performance of a broad range of hypothetical waste forms in generic repository concepts in clay, salt and granite.

Review of the available published safety assessments supports several qualitative conclusions about the likely impacts of hypothetical modifications to the waste inventory and waste form on the performance of various disposal concepts.

First, it is significant to note that all published safety assessments indicate that geologic disposal is safe. Estimated doses to humans are below regulatory limits, and are far below natural background, regardless of the disposal environment or waste form (spent nuclear fuel and/or high-level waste). Geologic disposal appears to be a robust concept that has the potential to provide safe isolation for a wide range of radioactive materials in a wide range of environments.

Second, thermal output of the waste affects loading strategies for all repository concepts, and peak temperature is generally treated as a design constraint that must not be exceeded. Lowering the thermal output of waste will allow more waste to be emplaced at higher density within a repository, but may have little impact on long-term performance other than the potential for a modest linear increase in estimated dose commensurate with any increase in the inventory of the dominant radionuclides contributing to dose.

Third, increases in the lifetime of specific waste forms have a potential to lower peak dose, or to delay its occurrence, if the waste forms stabilise the dominant radionuclides contributing to peak dose and if waste form lifetimes (e.g. expressed as a half-life based on a constant degradation rate) are long relative to the half-lives of the specific radionuclides or are a significant fraction of the regulatory period. In all cases, durable waste forms contribute to a multiple-barrier concept to provide defence-in-depth in isolating the disposed materials.

Fourth, removal of actinides from the waste form, e.g. by separation and transmutation, is likely to have little or no impact on estimated dose from repositories in reducing environments where actinides are relatively immobile, and could have a limited impact on estimated doses from repositories in oxidising environments where actinides are more mobile, lowering dose to the point where contributions from more mobile fission products (i.e.  $^{129}\text{I}$  and  $^{99}\text{Tc}$ ) become dominant. Increases in mobile fission products associated with higher burn-up of actinides may result in a modest linear increase in estimated dose, but overall performance is likely to remain well below any regulatory limits. More comprehensive separation and transmutation approaches that also remove long-lived fission products from the waste have the potential to lower estimated dose below the values published for current repository concepts.

### Thermal power and waste volume considerations

Analyses of the thermal behaviour of repositories begin with a consideration of the thermal output of the waste, typically expressed in terms of power per unit volume of waste ( $\text{W}/\text{m}^3$ ). However, for most long-term repository performance analyses, the thermal metric of interest is temperature as a function of time at various locations within the disposal region, which is a derived property dependent on multiple factors including thermal output of the radionuclides in the waste, density of those nuclides in the waste, design and operational considerations such as emplacement timing and geometry, and the physical properties of the host rock. A design constraint of keeping peak post-closure temperatures below a specified value (such as maintaining sub-boiling temperatures in a clay buffer surrounding waste packages) can be achieved through multiple approaches, including, for example, ageing the waste before emplacement, ventilating the emplacement areas before closure, decreasing the density of waste emplacement and decreasing the thermal output of the waste. Multiple approaches have been used in design concepts evaluated in existing safety assessments, and it does not appear that thermal constraints preclude implementation of any of the major disposal concepts for waste forms with a broad range of thermal output.

Modelling studies that hold all other aspects of repository design and operations constant while varying the thermal power of the waste and the spacing of waste packages, such as were done for the 2008 RED-IMPACT study [10], provide a useful insight into the potential benefits in efficient use of disposal volume. For example, model results suggest that waste from a full-recycle fuel cycle including separation of minor actinides could achieve sub-boiling temperatures in disposal galleries in clay and granite that are 30% to 40% of the length required for waste resulting from the generation of the equivalent electric power using an open fuel cycle (Table 7.1, Ref. [10]). Doubling the ageing time (to 100 years) for waste in which fission products provide the primary source of heat extends modelled reductions in required repository volume further, to a reduction factor of 13 in a hypothetical clay repository (Section 7.1.3, Ref. [10]).

The detailed safety assessments for direct disposal of spent fuel in the US, French and Swedish repository programmes [6, 7, 9] support a conclusion that regardless of the repository disposal concept and the properties of the waste stream, peak sub-surface temperature is a

constraint that can be met through engineering solutions without relying on separating heat-generating radionuclides from the waste stream. Overall estimates of long-term dose indicate that as long as temperature objectives are met in these analyses doses are well below regulatory limits, suggesting that radioactive decay heat itself is not a safety issue for disposal. Rather, management of radioactive decay heat should be seen as a design optimisation issue. As such, alternative fuel cycle options that affect thermal loading should be evaluated in system-level optimisation studies that address, among other factors, comparisons of the costs and benefits of licensing, constructing and operating processing facilities with the costs and benefits of licensing, mining and operating more or larger repositories that accommodate lower loading densities.

### Impacts of waste form lifetime on repository performance

In any disposal concept, the physical and chemical properties of the waste form have a potential to control the rate at which radionuclides leave the repository, and therefore have a potential to influence the time and magnitude of the estimated peak dose to individuals in the biosphere. Intuitively, the longer radionuclides remain within the waste form, the better the potential performance of the repository. In practice, however, repository models show that many other factors also influence the rate at which radionuclides may reach the biosphere, and the impacts of waste form lifetime itself need to be evaluated in the context of the full disposal system. Other potentially significant factors include water flux to and through the engineered barriers containing the waste form; degradation rates of the engineered barriers; chemistry of the water that contacts the engineered barriers and, after the barriers are breached, chemistry of the water that contacts the waste form; and radionuclide transport properties of the near- and far-field environments. Any one of these factors may dominate overall performance of the repository at various times in the future, and it is not always clear whether improved performance of an individual component will translate into significant improvements of the disposal system. For example, increasing the lifetime of a waste form by reducing its degradation rate may have little impact on the magnitude of the estimated peak dose if the primary barrier to release comes from another portion of the disposal system.

The safety assessment completed for the French clay site [7] provides an example of a disposal system in which overall performance is relatively insensitive to waste form lifetime because modelled releases are largely controlled by the slow rate of diffusive transport through the geosphere. For the reference case analyses, radionuclides are assumed to be released gradually from the matrix for spent fuel over approximately 50 000 years (p. 222, Ref. [7]). Possible sensitivity to this assumption was tested by assuming a ten-fold increase in dissolution rate of the fuel matrix, and time and magnitude of peak releases to the biosphere were essentially unchanged because the reference waste form lifetime of 50 000 years was already significantly shorter than the transport time through the geosphere (p. 325, Ref. [7]). Further increases in the fuel degradation rate (beyond the assumed factor of ten) would have no effect on overall release rate. Repository performance showed a somewhat greater sensitivity to increases in the degradation rate of high-level glass waste because its reference lifetime for the analysis was longer, “on the order of a few hundred thousand years” (p. 222, Ref. [7]). A sensitivity analysis examined the possible impact of an alternative conceptual model in which glass degradation rates remain constant in time rather than slowing when the surrounding water is saturated in silica, diminishing the waste form lifetime to thousands of years. Results for this alternative model showed an insignificant increase in the peak biosphere release of the dominant radionuclide contributing to dose,  $^{129}\text{I}$ , from  $8.6 \times 10^{-4}$  mol/yr to  $9.1 \times 10^{-4}$  mol/yr, but a potentially significant shift in the time at which peak release occurred, from 460 000 years to 250 000 years (Table 5.5-24, Ref. [7]). This temporal shift indicates that the relatively long lifetime of the glass waste in the reference analysis contributed effectively to delaying the peak release to the biosphere.

Results from the safety assessment for the Swedish granite repository proposed at Forsmark [9] provide similar insights. In the base case analyses for corrosion failure of waste packages (Section 10.6.5, Ref. [9]), spent fuel fractional dissolution rates in the reducing

environment range from  $10^{-6}$ /yr to  $10^{-8}$ /yr, corresponding roughly to waste form lifetimes ranging from 1 000 000 years to 100 000 000 years (Section 2.5.5, Ref. [11]). No releases occur in this analysis until after the first waste packages are breached, after approximately 500 000 years. Sensitivity analyses that vary the fuel dissolution rate (Figure 44, Ref. [9]) indicate that, within the range of values anticipated for reducing conditions, estimated dose to an exposed individual in the biosphere varies essentially linearly with the dissolution rate. At significantly higher dissolution rates such as might occur in oxidising environments (i.e. above  $10^{-5}$ /yr), total dose ceases to increase with increasingly rapid fuel degradation, presumably because dose is now limited by the rate at which nuclides are transported from the spent fuel to the biosphere, rather than by the rate at which they are released from the fuel.

While the degradation of the waste form may not be as important if the primary barrier to release comes from another portion of the disposal system, robust and durable waste forms are still desirable. Such waste forms in combination with the other multiple barriers add defence-in-depth to a disposal system concept, helping to increase the confidence of regulators and other stakeholders that the system would be protective of public health and safety.

### Impacts of actinide separations on repository performance

Understanding the potential for improvements in repository performance through the removal of specific radionuclides from the waste form by separation and transmutation processes requires understanding which radionuclides contribute the most to overall dose, which in turn requires consideration of the radionuclide transport characteristics of different disposal concepts. For repositories in oxidising environments, actinide elements will be relatively soluble and may dominate estimates of total dose. Removing actinides therefore has the potential to lower dose until the contribution from other radionuclides becomes the dominant contributor, at which point total dose will be insensitive to further reductions in the actinide inventory. For repositories in reducing environments, actinide elements will be relatively insoluble and are unlikely to dominate total dose, which will be largely dependent on the releases of highly mobile long-lived radionuclides, primarily  $^{129}\text{I}$ . Results of the published studies discussed here suggest that reductions in the actinide inventory for repositories in reducing environments may have little or no impact on overall performance estimates. More comprehensive separation and transmutation approaches that remove long-lived fission products as well as actinides from the waste have the potential to result in substantially lower estimates of long-term dose. Existing dose estimates are well below regulatory limits for all geologic disposal concepts that have been evaluated in detail, however, and the potential impacts on dose of separations of both actinides and fission products should be considered in the context of system-level analyses of the full fuel cycle.

Support for these observations can be found in each of the safety assessments considered here. Figure 1 shows estimated mean annual dose from individual radionuclides for the proposed Yucca Mountain repository (Figure 2.4-20b, Ref. [6]). Consistent with the relatively higher actinide solubilities in the oxidising environment, the largest contributors to the estimated peak mean annual dose are  $^{239}\text{Pu}$  (for the first 200 000 years, until radioactive decay lowers its significance),  $^{242}\text{Pu}$  and  $^{237}\text{Np}$ . In addition,  $^{226}\text{Ra}$ , which is a decay product derived from  $^{230}\text{Th}$  and  $^{234}\text{U}$ , makes a significant contribution to the total. Removal of the actinides from the waste form would result in an essentially linear decrease in dose from these four radionuclides. Removal of actinides would have no effect, however, on the contribution to total dose from the long-lived fission products  $^{99}\text{Tc}$  (a major contributor in the oxidising environment at Yucca Mountain during the first several hundred thousand years) and  $^{129}\text{I}$  (a major contributor at 1 000 000 years). Simplistically, the largest reduction in dose that could be anticipated by removing actinides from the Yucca Mountain inventory, assuming all other factors remain unchanged from their treatment in Ref. [6], is approximately a factor of 10, as shown in Figure 1.

Estimates of annual dose from the disposal of spent nuclear fuel in clay in the French Dossier 2005 analysis [7] indicate that in a reducing environment where transport occurs solely by diffusion, total dose is due entirely to the highly mobile species  $^{129}\text{I}$ ,  $^{36}\text{Cl}$  and  $^{79}\text{Se}$ , as shown in

**Figure 1: Estimated mean contributions from individual radionuclides to total mean annual dose resulting from the disposal of spent nuclear fuel and high-level glass waste in tuff**

Adapted from Figure 2.4-20b, Ref. [6]

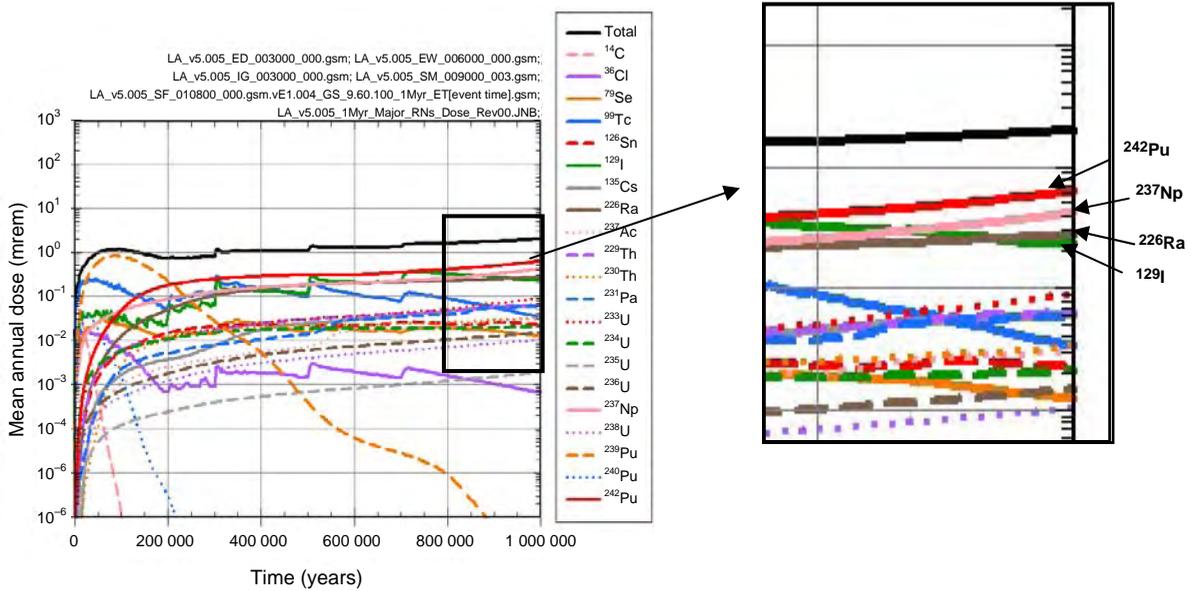
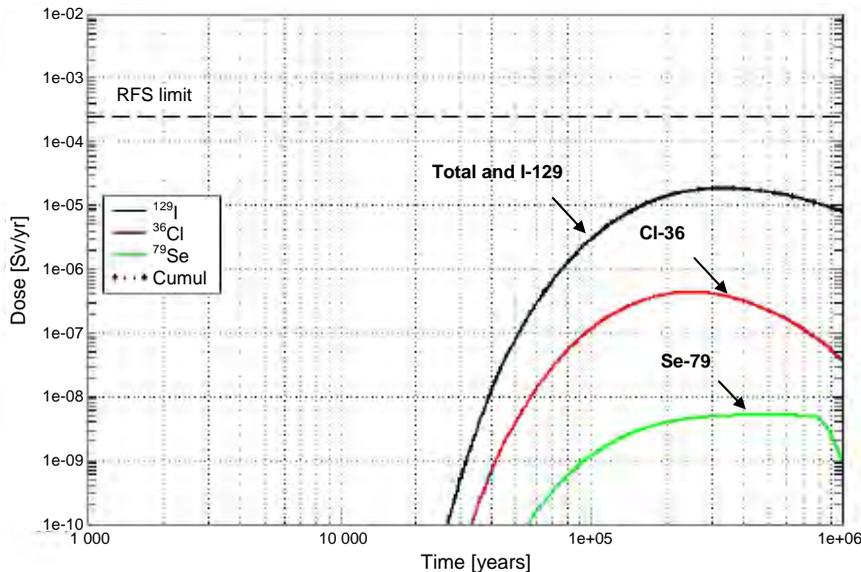


Figure 2 (from Figure 5.5-18, Ref. [7]). Actinides make no contribution to the total dose to humans because of their lack of mobility in the repository environment and the surrounding geosphere. Analyses performed for the disposal of spent nuclear fuel in the Opalinus clay in Switzerland provide additional confirmation of the potential effectiveness of the geosphere in containing actinides [8]. As shown in Figure 6.5-1 of Ref. [8], releases directly from the spent fuel into the near-field buffer are dominated by actinide species. Transport through the buffer significantly reduces the contribution of the actinides, and transport through the surrounding clay to the biosphere eliminates the contribution of the actinides to total dose.

**Figure 2: Major contributors to estimated annual dose resulting from the disposal of spent nuclear fuel in clay**

From Figure 5.5-18, Ref. [7]

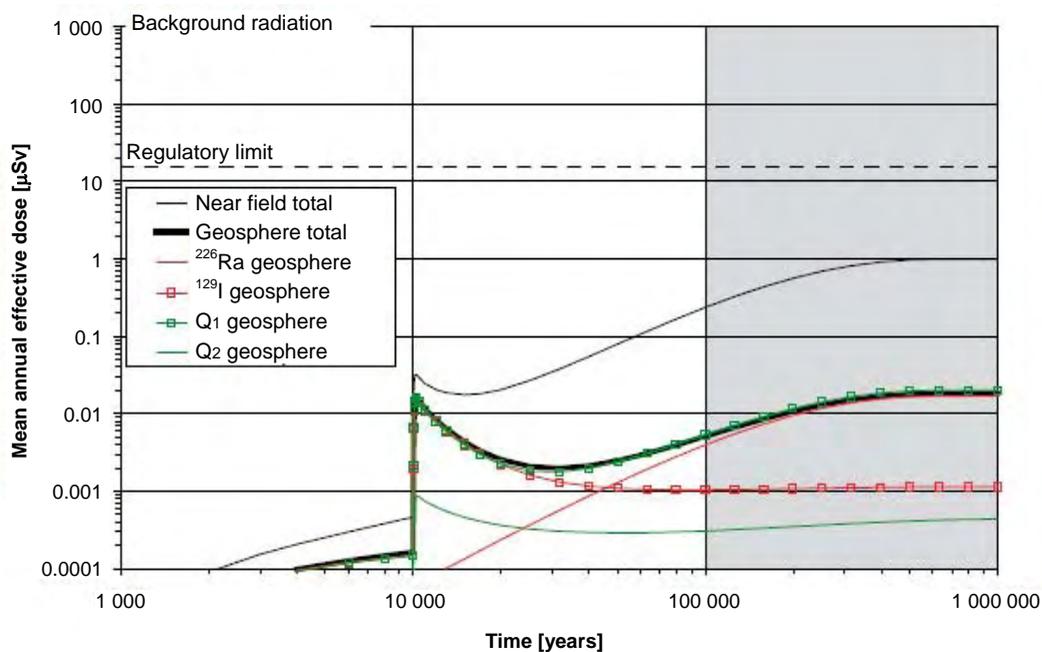


The safety assessment performed in Sweden for the disposal of spent nuclear fuel in granite at the Forsmark site shows similar results, with the addition of a major contribution from  $^{226}\text{Ra}$ , which is a decay product of  $^{234}\text{U}$  and  $^{230}\text{Th}$ , and is more mobile in reducing environments than its parent actinides (see Figure 3). In clay settings where radionuclide transport occurs by diffusion (e.g. Refs. [7, 8]), transport is sufficiently slow that  $^{226}\text{Ra}$ , with a half-life of approximately 1 600 years, decays before reaching the biosphere. In the case illustrated here, rapid transport is assumed to occur between the spent fuel and the biosphere by advection in fractures in the granite, and sufficient  $^{226}\text{Ra}$  reaches the biosphere to provide the primary contribution to the estimated peak annual dose.

**Figure 3: Major contributors to estimated annual dose resulting from disposal of spent nuclear fuel in granite**

Results shown here are based on an assumed pinhole failure of a waste package 10 000 years after disposal

Figure 10-18, Ref. [9]



## Conclusion

Results of published safety assessments for disposal of spent nuclear fuel and high-level radioactive waste provide insights into the potential impacts of alternative waste forms on the long-term performance of deep geologic repositories. Specifically, existing analyses suggest that modifications that reduce the thermal power of the waste or that reduce waste volume without increasing thermal loading have the potential to allow emplacing waste more densely in underground excavations. Costs and benefits associated with separating heat-generating radionuclides and managing and disposing of them separately can be compared to costs and benefits associated with more efficient use of repository volume in system-level analyses of the full nuclear fuel cycle. Modifications to waste forms that significantly decrease the rate at which the waste degrades have a potential to improve estimated performance of repositories if the resulting waste form lifetime is sufficiently long relative to the period of performance (e.g. on the order of hundreds of thousands of years), and if the waste form effectively immobilises those radionuclides that will be most mobile in the proposed geologic setting. Relatively smaller improvements in waste form lifetime, e.g. on the order of thousands or tens of thousands of years, are less likely to have a significant impact on the performance of a well-chosen and

well-designed repository because performance will be dominated by other, longer-lived components of the engineered barrier system and by slow transport through the geosphere. Results of published safety assessments suggest that modifications to the waste form that remove actinides that contribute most to the radiotoxicity of the waste (i.e. isotopes of plutonium, americium, neptunium and curium) could result in improvements in the estimated performance of repositories located in oxidising environments, where those species are relatively mobile. Removal of actinides alone (without separation of long-lived fission products) is less likely to result in an improvement in the estimated performance of repositories located in reducing environments where actinides are relatively immobile.

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## Relationship between geologic disposal characteristics and advanced nuclear fuel cycles with partitioning and transmutation

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### Abstract

*Advanced nuclear fuel cycles are currently being evaluated for their potential to improve on the nuclear fuel cycle characteristics and performance as exhibited by the current once-through nuclear fuel cycle, typically using LWR, with the goal of enabling continued and expanded use of nuclear energy. One of the areas being examined is the effect of using advanced fuel cycles incorporating partitioning and transmutation (P&T) on high-level and low-level waste management. The use of P&T alters the composition of high-level and low-level wastes along with other characteristics such as decay heat and mass. There is a wide range of technology options for partitioning of both actinides and fission products, and while active transmutation of actinides using neutrons may be a viable approach in either thermal or fast reactors along with other irradiation environments, options for fission product management are more related to using separate interim storage to allow for passive transmutation by radioactive decay of shorter-lived isotopes prior to disposal. There are also numerous potential geologic environments from which a suitable geologic disposal site may be selected, with many of these being intensively studied as part of national programmes on radioactive waste disposal. Each geologic environment has specific characteristics, such as the presence or absence of groundwater, the isolation of the site from the biosphere, the groundwater chemistry and the effects on corrosion as well as radionuclide transport, and the geologic stability. Radioactive waste disposal in each of these environments may also require the use of engineered barriers in addition to the isolation provided by the geology itself. It has been shown that the relative importance of actinides and fission products to the risk from geologic disposal varies from one geologic disposal environment to another. Based on the site-specific characteristics, the important isotopes and other desirable properties are identified and used to develop the requirements for potential advanced fuel cycles that would have wastes with properties that are favourable for disposal in that specific geologic environment. This paper presents the results of investigations into the synergism between the P&T that is specified for the advanced fuel cycle and the characteristics of the disposal environment. It identifies a range of potentially acceptable integrated nuclear fuel cycle and disposal systems, from those that require little P&T of used nuclear fuel to ones that benefit from extensive partitioning, transmutation of actinides and separate interim storage of fission products, considering the multiple characteristics of the radioactive wastes including potential radiation hazard and decay heat.*

## Introduction

Nuclear waste management includes all of the radioactive wastes associated with the nuclear fuel cycle, including wastes requiring geologic disposal such as used nuclear fuel (UNF) and certain long-lived highly-radioactive processing wastes, and low-level waste (LLW) that can be disposed with near-surface burial due to the much lower inherent hazard. Short-term storage prior to disposal can also be used as part of a nuclear fuel cycle option to manage very-short-lived radioactive materials, even those that are highly radioactive, allowing them to decay almost entirely and simplifying disposal. Alternative nuclear fuel cycles may also produce other hazardous and non-hazardous industrial wastes, depending on the processes used in the fuel cycle, although it is expected that such industrial wastes from nuclear fuel cycle activities would be handled as they are today for other industries, and would not present a significant additional burden due to the relative size of the nuclear energy infrastructure, even in cases where significant growth is assumed.

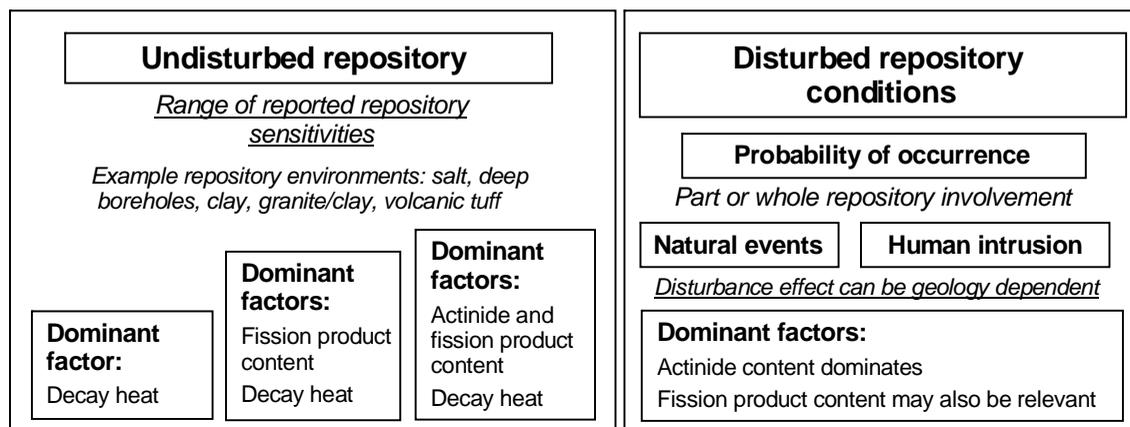
All use of nuclear power will generate radioactive wastes, whether the UNF itself is considered as waste, or if recycle is used, then the waste products from processing UNF, with waste disposal considered as an integral part of the fuel cycle. Contaminated equipment, wastes from operations and maintenance, and other wastes all contribute to the total waste production. The primary concern for radioactive waste disposal is the ability to provide the required isolation from the biosphere. For UNF or highly radioactive wastes, the consensus has been that deep geologic disposal is the preferred approach to provide the desired isolation of these materials from the biosphere, since many of the radioactive materials are hazardous for very long times, and only small amounts can pose a health risk. For wastes that are only slightly contaminated, the risk has been judged to be significantly less, and near-surface burial has been approved for such disposal. In both cases, other characteristics of the wastes may be important, such as volume or total mass, e.g. the mass of the waste form matrix as well as the mass of the disposed radionuclides. In reviewing studies on waste production, volume and total mass appear to be influenced by technology choices within a fuel cycle and appear to be amenable to waste reduction technologies or choice that can limit total waste mass and volume. In this paper, the potential for nuclear fuel cycles using partitioning and transmutation (P&T) to affect geologic disposal is explored, with alternative nuclear fuel cycles identified that may be used effectively.

## Repositories in deep geologic environments

The need for deep geologic disposal of used fuel and wastes that are highly radioactive has prompted investigation of a number of potential geologic environments in which to site a repository in order to evaluate the capability of the combined engineered repository and geologic environment to isolate these materials, such as the US programmes like the Nuclear Waste Terminal Storage Program starting in 1976, and activities created by the Nuclear Waste Policy Act of 1982, leading to the Yucca Mountain repository project [1]. An engineered repository includes all design features, such as disposal packages and any other engineered barriers to the release of radioactive materials, since acceptability of deep geologic disposal is likely to be determined by estimates of the peak dose rate for releases from the repository [2], which can be viewed as being determined by two factors: i) the radiotoxicity of the disposed inventory; ii) the degradation and transport characteristics of the engineered repository and the geologic environment. These two factors are analogous to “dose-response assessment” and “exposure pathway analysis” which are used for consequence evaluation in risk analysis [3]. Repository performance is the ability of the repository to provide the desired isolation.

Reviewing studies on the potential for geologic disposal, it is clear that there are several factors that may be important to the performance of the repository, including waste content and decay heat [4-10]. In considering the sensitivities of an engineered repository and the geologic environment affecting the ability to isolate the disposed materials, the analysis can be divided into two aspects, one for the performance under “nominal undisturbed conditions” of the repository, and the other considering the effects of disturbances, as shown in Figure 1, but both

**Figure 1: Factors related to engineered repository performance in a geologic environment for various geologic environments and for undisturbed and disturbed conditions**



aspects must be fully considered for an overall assessment of the proposed disposal system. Nominal undisturbed performance is where the conditions used for evaluating the geologic environment and designing the engineered repository are assumed to persist, while disturbed performance evaluates the effects of any disturbances that alter these conditions, either the engineered repository, the geologic environment or both. These two aspects can be considered either separately or combined into a single overall assessment for a repository, but both aspects need to be considered to arrive at an overall estimate of performance that would be required for licensing. For the purposes of understanding and evaluating the capabilities of an alternative nuclear fuel cycle to affect issues with deep geologic disposal, it is useful to consider undisturbed and disturbed performance separately, as summarised in Figure 1, mainly because it appears that nominal undisturbed performance may be predictable with comparatively lower uncertainty, while the probability of occurrence of disturbed conditions in the future is inherently uncertain, especially for human activities, and evaluating the consequences of the disturbance requires numerous assumptions about the nature and effects of the disturbance.

### ***Repository performance for nominal undisturbed conditions***

The performance of an engineered repository in a deep geologic environment under nominal undisturbed conditions assumes that the conditions used for evaluating and designing the repository remain unchanged during the time the repository is required to isolate the emplaced radioactive materials, i.e. the functional lifetime of the repository. From previous studies, there appear to be three groupings with different sensitivities to waste stream characteristics, as shown in Figure 1.

### ***Repository performance affected mainly by decay heat only***

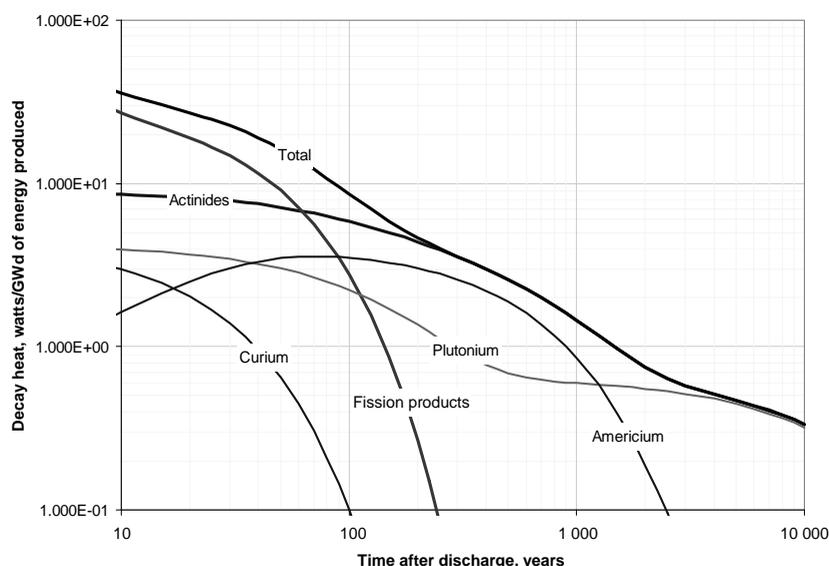
Repository options may exist where decay heat would be the dominant factor affecting repository performance for nominal undisturbed isolation performance, not being strongly sensitive to waste content since there may be essentially no releases to the biosphere. Examples may include:

- disposal in certain salt deposits, where temperature in the repository needs to be limited to prevent water migration and salt deformation that would facilitate releases;
- disposal in deep boreholes, where the temperature needs to be limited to prevent establishing a driving force for vertical convection that would facilitate releases.

For such deep geologic disposal options, the implication for an integrated fuel cycle appears to require limiting the volumetric decay heat generation rate, likely to levels much lower than is typical for UNF, and decay heat from both fission products and actinides can be important.

Decay heat generation in UNF decreases rapidly after discharge from the reactor, as does the decay heat for any wastes generated by processing the UNF. As shown in Figure 2 for UNF from an LWR with about 50 GWd/MTIHM discharge burn-up, the decay heat is dominated by fission products up to about 60 years after discharge. Decay heat from actinide elements such as plutonium and americium dominate after that time. The acceptable level of decay heat in a repository would be determined by detailed thermal analysis of the proposed engineered repository and the geologic environment, and could include the decay heat at time of disposal and the integrated decay heat from the time of repository closure until the peak temperatures in the repository are attained. This may require consideration of the importance of the decay heat from both actinide and fission product elements in the UNF or waste, although for some repository environments, only the shorter-term decay heat from fission products may be important. Another aspect is the time between discharge from the reactor and placement in the disposal environment. If shorter-term decay heat is important, interim storage of UNF or HLW for several decades or longer as part of a nuclear fuel cycle may be an effective means of reducing this short-term decay heat to levels acceptable for repository disposal.

**Figure 2: Decay heat from LWR UNF with 51 GWd/MTIHM discharge burn-up**



#### *Repository performance affected mainly by fission product content and decay heat*

There are some repository options where the nominal undisturbed repository isolation performance appears to be sensitive to both decay heat and the quantity and specific inventory of fission products, as some fission products may have potential release pathways. Examples may include:

- disposal in thick clay layers with saturated reducing conditions;
- disposal in a hard rock environment using clay with saturated reducing conditions surrounding disposal packages, as for a repository in granite.

In these cases, the fission product content affects the peak dose rate for individual exposures. Dominant fission product isotopes may include  $^{135}\text{Cs}$ ,  $^{129}\text{I}$ ,  $^{126}\text{Sn}$ ,  $^{99}\text{Tc}$ ,  $^{79}\text{Se}$ ,  $^{36}\text{Cl}$  and  $^{14}\text{C}$ . Most of the fission products are not amenable to recycle for transmutation, but some can be recycled such as  $^{129}\text{I}$  and  $^{99}\text{Tc}$ . As with the previous case, decay heat is an important parameter, affecting both repository performance and use of repository space. Both actinide elements and fission products can be important for decay heat.

*Repository performance affected by actinide content, fission product content and decay heat*

There are some repository options where both actinide and fission product content can be important to the nominal undisturbed repository isolation performance, as well as decay heat, as release pathways for radionuclides might develop. One example may include the disposal in volcanic tuff in an unsaturated oxidising environment.

Both actinides and fission products can be important to the peak dose rate in these cases. Decay heat is an important parameter for affecting repository performance and use of repository space in this case as well, with both actinides and fission products contributing to the decay heat.

The differentiation of waste disposal impacts between actinides and fission products can be important for the fuel cycle option because these two categories of materials (fission products and actinides) have very different characteristics in neutron irradiation, where some actinide isotopes can be actively involved in fission and energy production while fission products mainly absorb neutrons.

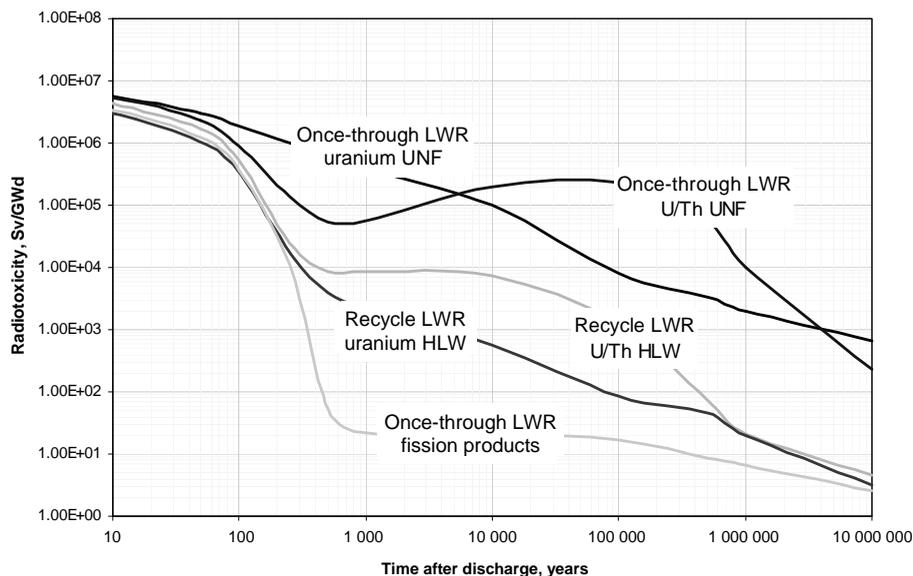
**Repository performance in response to disturbances**

In reviewing the studies on repository performance, it is seen that disturbances to a geologic repository can be important in evaluating overall performance, since all repositories may have releases to the biosphere as a result, including some cases where overall repository performance is dominated by the effects of disturbances. Disturbances can be caused by human activities or by natural events. Human intrusion includes exploratory drilling and mining, and examples of natural events are igneous and seismic episodes. In all cases, the repository environment is disturbed from the nominal undisturbed conditions that were assumed for the development of the repository, with implications for the subsequent ability of the repository to provide the desired isolation of the disposed materials.

The effects of human intrusion appear to vary significantly with the geologic environment. For example, a repository in salt could be vulnerable to exploratory drilling, resulting in penetration of the salt and surrounding layers, with the potential for allowing water into the repository. In such a case, all of the disposed materials may eventually be at risk of release due to the resulting corrosion of disposal packages and contents, although there is also the risk of direct exposure from the contents of the disposal package that was breached. On the other hand, drilling through a repository in volcanic tuff or clay might only affect the waste package encountered with no further consequences, since water is already present in those environments. The previous studies indicate that it may not be possible to provide a scientific estimate of the probability of human intrusion, although it is recognised that the likelihood of intrusion may be reduced by appropriate selection of the repository site away from known natural resources. Since the contents of one or more waste packages would be at risk, both the actinide and fission product elements contributing to the radiotoxicity could be important to the peak dose rate associated with these events, as shown in Figure 3 for ingestion radiotoxicity. The radiotoxicity is plotted for both uranium-based and thorium-based (Th/<sup>233</sup>U) fuel used in an LWR on a per GWd basis for both once-through and recycle approaches. As can be seen in the figure, the radiotoxicity for UNF from LWR is generally similar whether uranium-based or Th/<sup>233</sup>U fuel is used. Also, the fission product contribution is shown, which would be the lower limit for reducing ingestion radiotoxicity when actinide recycle is used, i.e. there are no losses of actinides to the wastes.

Natural events causing disturbances are large-scale phenomena that conceivably affect all of the disposal packages in the repository. Once the packages are breached, the entire inventory is available for release and transport. Depending on the assumptions made about the exposure pathway, again both dominant actinide and fission product elements contributing to the radiotoxicity can be important to the peak dose rate for such events. Predicting the probability of future natural events is also uncertain, although past history may be usable as an approximate indicator of occurrence probability.

**Figure 3: Ingestion radiotoxicity for uranium-based and thorium-based fuel used in an LWR for both once-through (UNF) and recycle (HLW) options**



An important implication for fuel cycle options from the preceding discussion is that there does not appear to be a single, simple or universal correlation between fuel cycle waste characteristics and disposal challenges. The importance of waste stream characteristics depends on the details of the disposal host environment and the repository design, which even then can be altered by potential disruptive processes.

### Technical implications for a nuclear fuel cycle option

Based on the published information reviewed for this study, the technical implications for an integrated fuel cycle as a result of specific geologic environments depend on the relative importance of nominal undisturbed performance and the effect of disturbances. In all cases, it appears that uncertainties about the repository environment and exposure pathways may remain large, in part due to the fundamental complexity of a geologic environment and spatial variations in characteristics, although some environments may have lower uncertainties than others, and mainly due to the imprecise nature of prediction of future events, natural or man-made. As a result, there is no single, simple answer for what a nuclear fuel cycle option needs to do to benefit deep geologic disposal, aside from the fundamental principle that the less hazardous material placed in the environment, the better.

### ***Geologic disposal – repository performance dominated by nominal undisturbed conditions***

Using Figure 1, fuel cycle implications for undisturbed geologic disposal are as follows:

- Decay heat is the dominant parameter affecting repository performance:
  - The decay heat of the fission products can be mitigated or eliminated with interim storage. Alternatively, an integrated fuel cycle could use separations to isolate high decay heat fission products from the remaining wastes, storing only those fission products for an extended period of time. Fission product recycle and transmutation is not likely to be effective.
  - The decay heat from the actinides persists for longer times, at least a thousand years, and is addressable by separation and recycle of the actinides, very long-term storage (many hundreds of years or longer) or substantial reduction in the volumetric heat rate.

- Fission product content and decay heat are the dominant parameters:
  - Recycle of fission products is not likely to be effective for reducing fission product content, as fission products are not amenable to transmutation by neutron irradiation.
  - Decay heat is more an operational and engineering issue in this case, and both fission products and actinides may be important in reducing decay heat. Interim storage may be effective for the fission products, while recycle of the actinides would be required.
- Actinide content, fission product content and decay heat are all important parameters:
  - Recycle of the actinide elements can be done effectively with recycle.
  - Decay heat is more an operational and engineering issue in this case, and both fission products and actinides may be important in reducing decay heat. Interim storage may be effective for the fission products, while recycle of the actinides would be required.

As discussed earlier, the distinction between fission product and actinide impacts is important to fuel cycle options because of the differing characteristics of these materials in fuel cycle technologies.

### ***Geologic disposal – performance dominated by nominal disturbed conditions***

If the effects of disturbances dominate the overall performance of the repository, uncertainty will be very high, in proportion to the ability to predict such future events. For the two types of disturbances, human and natural, the importance of the disposed material characteristics may vary considerably.

- Human intrusion:
  - For human intrusion into a repository where no groundwater is present for undisturbed conditions, such as by drilling into the repository, the effects can be significant, especially if water is introduced as a result. It would seem that the only approach to reducing the resulting peak dose rate from such a failure of the repository boundary is to lower the radiotoxicity of the disposed inventory either by restricting the inventory or by separating and recycling the dominant peak dose rate contributors. Both actinides and fission products may be important, according to the radiotoxicity shown in Figure 3, depending on the assumptions and characteristics of the resulting exposure pathways.
  - For human intrusion into a repository where groundwater is always present, the effects of human intrusion, as reported in studies to date, appear to be relatively minor. An alternative fuel cycle may have little or no effect in this case, depending on the projected disposal package contents.
- Natural events:
  - The effects of large-scale natural disturbances, such as seismic and igneous events, may be significant since the disturbed environment may not have the same isolation capability, especially in cases where the disturbance introduces water into a repository that had essentially no water. In all of these cases, the scale of the disturbance is such that studies assumed that the entire inventory of the repository may be at risk and released from the repository environment. For example, in the case of Yucca Mountain, the dose rate for the igneous intrusion event is dominated by isotopes of the actinide elements [7]. Both actinide and fission product elements can be important for such conditions, and reducing the dose rate resulting from such disturbances would require lowering the radionuclide inventory.

Overall, the consequences of disturbances can be reduced by lowering the inventory in the repository, which may have the effect of lowering the importance of the disturbance events to the overall performance of the repository, increasing confidence that the repository provides the desired isolation.

### ***Performance determined by both nominal undisturbed and disturbed conditions***

The studies that have been reviewed demonstrate that both nominal undisturbed conditions and disturbed conditions would likely be considered in developing the performance assessment of geologic disposal. For any alternative fuel cycle, the geologic disposal environment and any associated engineered systems should be considered as part of the integrated fuel cycle since it will determine what, if any, impact the alternative fuel cycle would have on waste management. For identifying those fuel cycles that may be able to make a significant difference to the current nuclear waste management situation, measures to address both undisturbed and disturbed conditions are considered.

### **Fuel cycle options**

Given the technical implications summarised above, the effectiveness of using nuclear fuel cycles that include P&T on geologic disposal can be described. Considering the transmutation aspect of P&T, it is important to consider the nature of nuclear fission and its inherent limitation. Nuclear fuel can only be used as long there is sufficient fissile material to support the nuclear reaction, considering that the fission product materials that absorb neutrons, “poisons”, build up as the fuel is consumed. Since these fission products are not removed with a once-through fuel cycle, the fission process eventually stops, limiting the use of the fuel. The UNF is then disposed using deep geologic disposal. As a consequence, any use of fuel in a reactor with a once-through fuel cycle requires that a relatively large quantity of nuclear fuel be provided for power production, and that all of the UNF be buried at a geologic disposal site. Typically, in a once-through system the fission products build nearly linearly with fuel burn-up and actinide content approaches (but does not reach) a quasi-equilibrium level with ongoing isotopic evolution with increasing burn-up. This combination limits the potential waste management benefit for once-through systems to only modest improvements. An exception to this could be an externally driven system with sufficient neutrons to approach nearly complete burn-up, where the final actinide content could be lowered.

With recycle, actinides are typically separated and reused in fuel, producing further energy and fission products. By so doing, on the basis of energy generated, the amount of highly hazardous material per unit of energy produced that would need to be buried using geologic disposal can be greatly reduced. This reduction could be used to greatly lower the inventory to be disposed of in a given repository site, reducing potential releases and making uncertainties less important. Alternatively, the greatly lower decay heat and radionuclide inventory could permit a disposal site to effectively accept high-level waste associated with a near 100-fold increase in energy generation. However, it is noted that even with actinide recycle where all UNF is processed, there would still be need for a disposal site for the burial of the fission products and the radioactive material losses during reprocessing.

Options to significantly improve nuclear waste management focus on those materials requiring deep geologic disposal, and potential options are generally based on greatly reducing the waste inventory of the radionuclides important for radiation exposure and decay heat. While the specific approach that may benefit undisturbed repository performance will be dependent on the engineered repository and the specific geologic environment, actinide elements can be important for both radiation exposure and decay heat, and long-lived fission products can be important for radiation exposure. Whether a benefit would be obtained or not for a specific deep geologic disposal option cannot be known until both a host environment and repository design are known, and even then the resulting analyses are at least partly subjective due to the assumptions that need to be made. Typically however, for undisturbed conditions, important fuel cycle waste characteristics include at least decay heat limitation, and may include fission product and actinide inventory. For disturbed conditions, both actinides and fission product inventories are likely to be important.

The following general categories of both uranium-based and thorium/fissile-based fuel cycles will contain one or more options that may be beneficial:

- Recycle options with all UNF processed, recycle of actinide elements:
  - Recycle: Critical – thermal neutron systems;
  - Recycle: Critical – fast neutron systems;
  - Recycle: Critical – thermal and fast neutron systems;
  - Recycle: Subcritical – thermal neutron systems;
  - Recycle: Subcritical – fast neutron systems;
  - Recycle: Subcritical – thermal and fast neutron systems;
  - Recycle: Critical and subcritical – thermal neutron systems;
  - Recycle: Critical and subcritical – fast neutron systems;
  - Recycle: Critical and subcritical – thermal and fast.
- Once-through: Subcritical – fast neutron systems, essentially complete fuel consumption.

The key aspects of the recycle fuel cycles are that all UNF needs to be processed, since UNF actinide content is large compared to processing and fuel fabrication losses, dominating disposal inventory, and that all of the actinides should be recycled, not just uranium and plutonium.

## Summary

Achieving significant benefits to nuclear waste management involves addressing deep geologic disposal, LLW disposal and storage needs. Regarding deep geologic disposal, it was observed that the isolation performance of each combined engineered repository and geologic environment was not the same, with some being more sensitive to actinide elements as compared to fission products, and some being more sensitive to disturbances to the repository than others. With respect to disturbances, it appeared that the risk from disposal may be reduced by reducing the actinide inventory in the repository to reduce both actinide elements and their decay products (an action important for either uranium or thorium use), although in some cases it would be beneficial to reduce the inventory of certain fission products as well. While this could be accomplished by reducing the planned capacity of a repository, either complete consumption of the fuel or recycle of the actinide elements was seen to provide a significant benefit. However, it was also observed that there is subjectivity in the analysis of both undisturbed and disturbed events, especially for the assumptions made as to the nature and consequences of disturbances, and uncertainty is high for predicting future events. Reduction in inventory lessens the importance of these uncertainties in an overall assessment of the repository capabilities.

The situation is more complex for undisturbed repository performance. Some repositories may not have releases for such conditions, but achieving this may be difficult in practice and may require substantial reductions in the decay heat of the emplaced materials to enable such disposal. For the first few decades, decay heat is dominated by short-lived fission products, and this could be addressed by using interim storage prior to disposal. However, the longer-term decay heat, out to two thousand years or more, resulting from some of the actinide elements can only be addressed by recycle to keep these elements out of the wastes, aside from minor processing losses, or by distributing the waste over larger emplacement areas, which might also require fuel separation to reduce actinide concentrations to levels below that of intact UNF. Other repositories are estimated to have very small losses for undisturbed conditions, with some preferentially releasing actinide elements and other releasing fission products. For repositories sensitive to actinide elements, recycle can have a significant impact on such releases. However, recycle does not appear to be effective for those sensitive to fission products, since most fission

products are not amenable to recycle. It was observed that analysis of undisturbed performance may also be partly subjective due to assumptions that are made, but in general there appeared to be less uncertainty about performance for undisturbed conditions.

Overall, for a nuclear fuel cycle option to reduce the risk from deep geologic disposal, actinide recycle appears to benefit disturbed performance for all repositories, and undisturbed performance for some repositories. Actinide recycle may also enable disposal in some types of repositories by reducing long-term decay heat. Very few fission products are amenable to recycle, so fission product recycle may have limited benefit.

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## The environmental impacts of Korean advanced nuclear fuel cycle KIEP-21 and disposal concepts

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### Abstract

*We have performed a performance assessment to investigate effects of waste forms and repository designs by comparing the case of direct disposal of used PWR fuel in the Korean Reference Repository System (KRS) concept with the case of Advanced Korean Reference Disposal System (A-KRS) repository containing ILW and HLW from the KIEP-21 system. Numerical evaluations have been made for release rates of actinide and fission product isotopes at the boundaries of the engineered barrier system (EBS) and the natural barrier system (NBS) by the TTB code developed at UC Berkeley. Results show that in both cases, most actinides and their daughters remain as precipitates in the EBS because of their assumed low solubilities. The radionuclides that reach the 1 000-m location in NBS are fission products,  $^{129}\text{I}$ ,  $^{79}\text{Se}$  and  $^{36}\text{Cl}$ . They have high solubilities and weak or no sorption with the EBS materials or with the host rock, and are released congruently with waste form alteration. In case of direct disposal, a contribution of 2% of iodine is assumed to be accumulated in the gap between the cladding and fuel pellets released after failure of the waste package and cladding dominates the total release rate. With increase in the waste form alteration time, the peak value of total release rate decreases proportionally because the dominant radionuclides are fission product isotopes, which are released from waste forms congruently with waste form dissolution. It has been shown by PHREEQC simulation that actinide solubilities can be significantly affected by pore water chemistry determined by the evolving EBS materials, waste forms and compositions of groundwater from the far field.*

## Introduction

Korea Atomic Energy Research Institute (KAERI) has been developing pyroprocessing technologies called Korean, Innovative, Environment Friendly and Proliferation-resistant System for the 21<sup>st</sup> Century (KIEP-21) to continuously utilise the benefits of nuclear energy while minimising the burden of used fuel arising [1]. The pyroprocessing technique for KIEP-21 is intended to recover more than 99% of the actinide elements included in used PWR fuel, and thus to minimise the heat load and volume of waste. Correspondingly, KAERI has also been developing the Advanced Korean Reference Disposal System (A-KRS) to accommodate radioactive wastes, which are to be generated from the KIEP-21 system [2].

In this study, we have performed an analysis to investigate effects of waste forms and repository designs by comparing the performance of direct disposal of used PWR fuel in the Korean Reference Repository System (KRS) concept with the performance of A-KRS repository containing intermediate-level wastes (ILW) and high-level wastes (HLW) from the KIEP-21 system [2, 3]. To compare performances, numerical evaluations have been made for the release rates of actinide and fission product isotopes at the boundaries of the engineered barrier system (EBS) and at a natural barrier system (NBS) with the Transport to Biosphere (TTB) code developed at UC Berkeley [4]. For a detailed description of the TTB model and mathematical formulations, refer to the previous paper [4].

We have studied solubility effect on radionuclide transport. We evaluated actinide solubilities in different geochemical conditions such as granitic groundwater, cement water, bentonite pore water and monazite pore water using the PHREEQC (V.2.17) code [5], based on hydro-geochemical and hydro-geological data obtained at KAERI Underground Research Tunnel (KURT) and reference papers [6].

## Waste forms and repository concepts

From the pyroprocessing of the used PWR oxide fuel, the provisional five waste streams are generated and solidified into: i) metal waste-ILW; ii) ceramic waste-HLW; iii) vitrified waste-HLW; iv) ceramic waste-ILW; v) vitrified waste-ILW (Figure 1) [7], also summarised in this section. For repository concepts, we compare KRS for direct disposal of used nuclear fuel, with A-KRS for the provisional five wastes from the KIEP-21 system. In this analysis, we assume disposal of 26 000 MTU of used PWR fuels or equivalent for the KIEP-21.

### Five waste forms from KIEP-21

#### *Metal waste (Waste Stream 1)-ILW*

Metal wastes contain effluents from two different processes: i) cladding hulls from the chopping and decladding process; ii) insoluble noble metal fission products from the electrorefiner [1]. The cladding hulls are rinsed to remove the adhered fuel and FP materials before being melted to a corrosion-resistant metal alloy. The fissile materials and FP (except for noble metals) remaining in a hull are dissolved in a LiCl-KCl salt using zirconium chloride. The noble metal fission products contain a small amount of actinides left behind in the anode basket after the electrorefining process. They are also rinsed to remove the actinide elements in the form of chlorides. The rinsed actinide and fission product chlorides are returned to the electrorefiner, while the residual metallic fission products are melted together with the cladding hull and an additional stainless steel to produce a corrosion-resistant metal alloy.

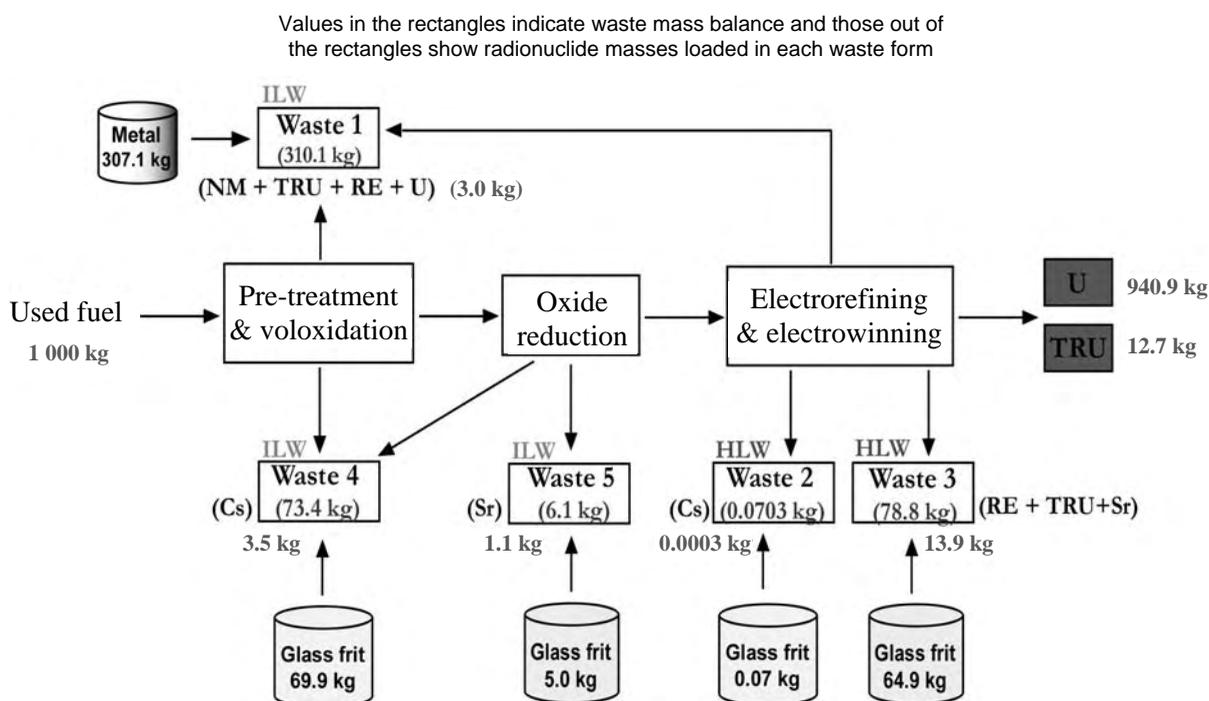
#### *Ceramic waste (Waste Stream 2)-HLW*

The LiCl-KCl waste salt from the electrowinning process contains a very small amount of <sup>135</sup>Cs and <sup>137</sup>Cs. In this study, we neglect this waste stream because of a small amount compared with other streams.

### Vitrified waste (Waste Stream 3)-HLW

The LiCl-KCl waste salt from the electrowinning process contains a considerable amount of rare-earth elements and a small amount of transuranic (TRU) elements and Sr. They are precipitated into their oxide or oxychloride forms via reactions with oxygen gas. When the precipitates are fully settled, the upper layer that is mainly composed of LiCl-KCl salt is separated from the precipitate part containing the rare-earth elements. The remaining salt in the precipitate phase, which is a mixture of the precipitates and eutectic salt residue, is separated and recovered from the precipitates using a vacuum distillation/condensation method. Finally, the remaining rare-earth precipitates are converted into stable oxides by simultaneous de-chlorination and oxidation reactions. These are then vitrified with borosilicate glass.

**Figure 1: Flow sheet for treatment of 1 MTHM of used PWR fuel with 4.5 wt.%  $^{235}\text{U}$ , 45 000 MWd/MTU and five provisional waste streams**



### Ceramic waste (Waste Stream 4)-ILW

The LiCl waste salt from the electrolytic reduction process contains Sr and Cs. After recovering and separating strontium by using  $\text{Li}_2\text{CO}_3$  in LiCl waste salt, a small amount of residual Cs is removed from the LiCl waste salt by applying an ion-exchange process with an inorganic material such as zeolite. In addition, 98% of Cs released from the voloxidation process is captured by a fly-ash medium at 650°C. Both  $^{135}\text{Cs}$  and  $^{137}\text{Cs}$  from the voloxidation process and the waste LiCl salt are fabricated to a ceramic waste form with an addition of a solidification agent such as glass frit. This waste form also contains  $^{129}\text{I}$  and  $^{14}\text{C}$  as well as  $^{135}\text{Cs}$  and  $^{137}\text{Cs}$ .

### Vitrified waste (Waste Stream 5)-ILW

Strontium included in the waste LiCl salt from the electrolytic reduction process is solidified by vitrification. First, Sr is precipitated into the form of a carbonate ( $\text{SrCO}_3$ ) by addition of  $\text{Li}_2\text{CO}_3$ . Then,  $\text{SrCO}_3$  separated from the molten LiCl salt is converted into its oxide form ( $\text{SrO}$ ) through a thermal decomposition. Finally, SrO is fabricated to a vitrified waste form.

## **Repository concepts**

The KRS is similar to the Swedish KBS-3 repository concept. The A-KRS concept is a combination of interim storage and a final repository, and the system is constructed at two separate depths: 200 meters and 500 meters. Basically, the HLW is to be finally disposed of at the 500-m level, while the ILW is disposed of at the 200-m level. The 200-m level space would also be used as the interim storage for wastes with high decay heat emission. The wastes containing mostly short-lived radionuclides such as  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  should be stored and disposed of at the 200-m level. Used PWR fuels would be temporarily stored in the tunnels at the 200-m level before they are processed by the KIEP-21 system. In Ref. [2], it is assumed that the disposal tunnels remain open for at least 100 years before final closure, although a detail scheme has yet to be determined.

KAERI proposed three layout options, one of which is adapted for this study. Tunnels with dimensions 150 m long, 8 m wide and 8 m high are considered. Eight hundred fifty-five (855) Compact-metal Disposal Packages (C-MDP) of Waste 1 (metal) in one tunnel, 2 600 storage racks (SR) of Waste 4 (ceramic) in nine tunnels and 2 600 SR of Waste 5 (vitrified) in two tunnels are finally disposed of at the 200 m level. HLW 2 (ceramic) and 1 300 storage and disposal containers (SNDIC) of HLW 3 (vitrified) in 16 tunnels are disposed of at the 500 m level. For detailed descriptions of the waste package and concept of package array in a disposal tunnel, refer to the paper [8].

The scheme before final closure, i.e. how the interim storage and the final disposal are combined and managed is the central matter of consideration, because it is related to retrievability/reversibility of the entire system, and because it determines the basis of comparison for repository footprint and cost between two concepts. It also relates to a question of inter-generational equity and fairness, because the A-KRS concept assumes a long cooling time. These need to be discussed at depth for comprehensive comparison, and will be further investigated in the present project.

## **Comparative and parametric studies**

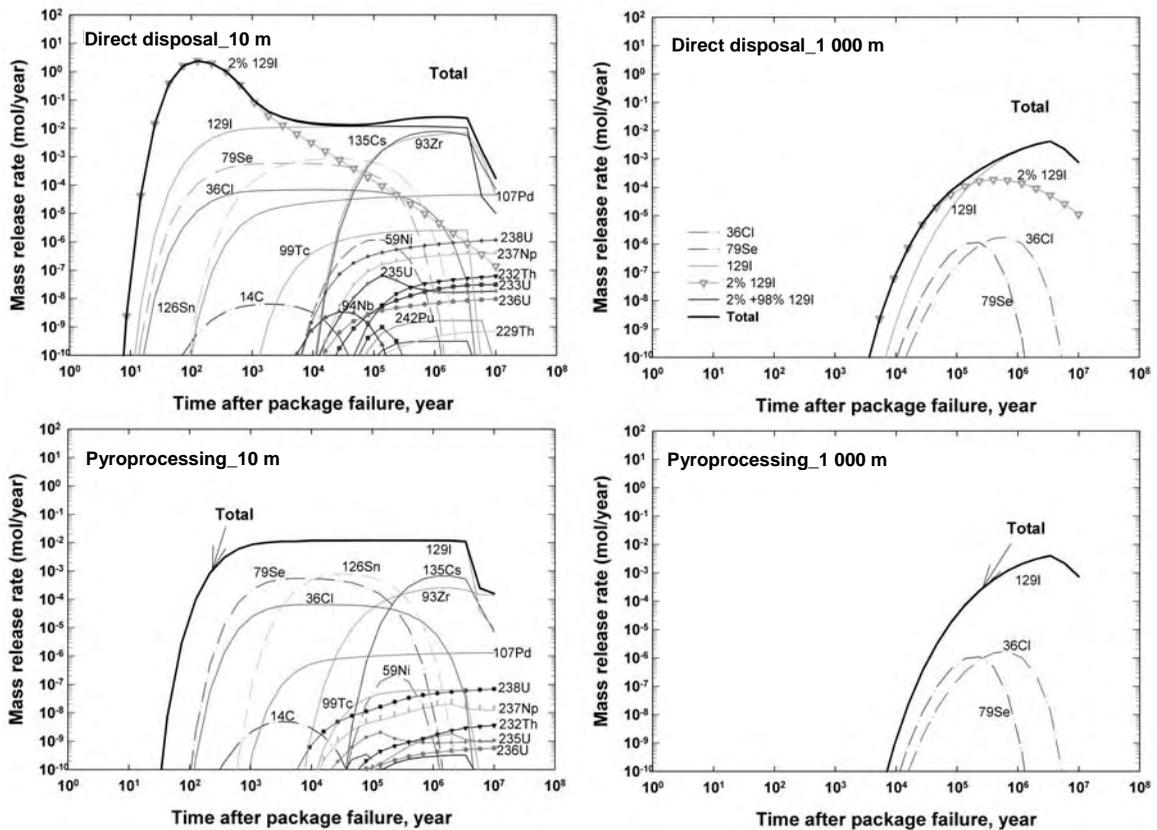
As comparative and parametric studies, we have considered the effects of schemes in the back-end fuel cycle, waste form alteration time ( $T_1$ ) and radionuclide solubility. Values of 4 000, 40 000, 400 000 and 4 million years for  $T_1$  are assumed. The radionuclide release by the waste form alteration is assumed to begin at the package failure time point.

## **Effects of back-end fuel cycle schemes**

We compared the total performance of multiple waste forms [2, 8] from the pyroprocessing in KIEP-21 disposed of in the A-KRS with that of used PWR fuel disposed of in the KRS concept [3]. Total mass release rates of radionuclides have been compared at the EBS surface, and in the far-field of geological formation on the same original fuel mass (26 000 MTU) basis. To get the total mass release rate of radionuclides for different waste forms from pyroprocessing, the result of the mass release rate from each single waste package was multiplied by the number of packages, 855, 1 300, 2 600 and 2 600 for Waste 1, Waste 3, Waste 4 and Waste 5, respectively, and summed. In case of direct disposal, the mass release rate of a radionuclide resulting from a single waste canister has been multiplied by 14 788, which is the total number of canisters for 26 000 MTU. The results are compared in Figure 2. Note that uranium and TRU elements are separated for future recycle and only small fractions of these are included in waste forms from the pyroprocessing case, while all nuclides are contained in one type of canister for the used PWR fuel case. The assumed conditions for the waste form alteration are the same, except that 2% of iodine accumulated in the gap between the cladding and fuel pellets after failure of the waste package and cladding is calculated separately for the direct disposal case.

Because of the solubility limit mechanism, only a small amount of the actinides can actually be released from the EBS to the surrounding NBS as shown in either case. Before released from the outer boundary of the EBS, most actinide radionuclides have decayed out, while most

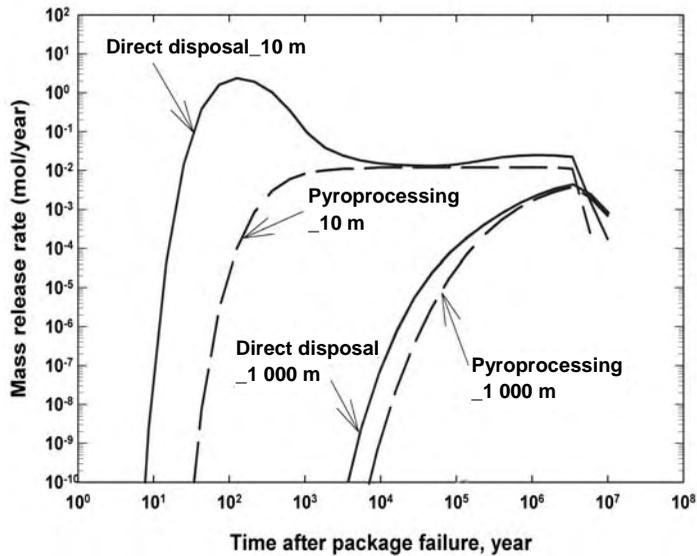
Figure 2: Total mass release rates of radionuclides at the 10-m and 1 000-m locations in NBS for the pyroprocessing wastes and the used PWR fuels with the waste form alteration time of 4 000 000 years



congruently-released fission product radionuclides other than short-lived radionuclides survive. The contribution from iodine, both in the gap and in the fuel matrix, dominates the total release rate for the direct disposal case at the 10-m location. The other major contributors at the 10-m location are  $^{79}\text{Se}$ ,  $^{36}\text{Cl}$ ,  $^{126}\text{Sn}$ ,  $^{135}\text{Cs}$  and  $^{93}\text{Zr}$ . In the pyroprocessing case, because iodine has already been separated and solidified into a dedicated waste form, such a pulse as observed in the used fuel case is not observed. Because 99% of actinides are recovered, the boundary conditions determined by the TTB code indicate that they are released congruently with alteration of waste forms, and so generally magnitude of peaks for actinides is reduced compared to direct disposal of the used PWR case. The major contributors at the 10-m location of pyroprocessing case are also  $^{129}\text{I}$ ,  $^{79}\text{Se}$ ,  $^{36}\text{Cl}$ ,  $^{126}\text{Sn}$ ,  $^{135}\text{Cs}$  and  $^{93}\text{Zr}$ . While the result of the total mass release rate of radionuclides for direct disposal case also includes the 2% gap-inventory effect of iodine, its pulse-like contribution is not observed for the 1 000-m location case. This is because of the matrix diffusion assumed in the transport through the fractured geological medium in the far field. In this model, the rock matrix acts as a reservoir to delay and spread arrival of radionuclides at the observation point. Therefore the result of the total mass release rate of radionuclides for pyroprocessing is almost the same as the direct disposal case. The major contributors at the 1 000-m location are  $^{129}\text{I}$ ,  $^{79}\text{Se}$  and  $^{36}\text{Cl}$  for both disposal concepts.

Figure 3 shows the total mass release rates of all radionuclides at the 10-m and 1 000-m locations for the pyroprocessing wastes and the used PWR fuels. At the 10-m location, two cases show a clear difference. This is considered to be due to effects of source conditions, including the waste forms and the engineered-barrier configuration. The main factor that gives smaller release rate for the pyroprocessing case is better solidification of iodine. This result indicates that the pyroprocessing coupled with waste forms suitable for confining each radionuclide can significantly improve the engineered-barrier performance. At the 1 000-m location, it is observed

**Figure 3: Summary of total mass release rates of all radionuclides at 10-m and 1 000-m locations for the pyroprocessing wastes and the used PWR fuels**



that the main contributors are those three fission product isotopes, and the two cases compared here do not display any apparent difference. This comparison indicates that the performance gain observed at the near field may not be so important for the final repository performance, but it is worth noting that the present analysis does not include detailed repository layout conditions or geological conditions in the far field; to draw more decisive conclusions, site-specific information must be known. Until such detailed information has become readily available, the results such as shown in this study should be understood in a stylised manner.

We compared the volume and heat emission of wastes from these two cases. The volume of waste from KRS is 3.06 m<sup>3</sup>/tonne and that from A-KRS is 0.37 m<sup>3</sup>/tonne. About 1/8 of waste volume reduction can be achieved. It should be noted that the volume of wastes after the pyroprocessing in various waste forms is determined not only by heat emission rates but also by various chemical durability considerations. The values used in the present comparison are still preliminary, and the volume of separated uranium and TRU is not taken into consideration for the pyroprocessing case.

The heat emission rate of the ILW and HLW from the KIEP-21 system after a 300-year cooling time has been estimated by the ORIGEN code to be 2.00 watts/tonne-spent fuel equivalent while 134 watts/tonne is emitted from the used fuel with the same cooling time. The difference comes from the fact that most of the TRU isotopes are recovered from the used PWR fuel through pyroprocessing, while the same amount of fission products are included for both cases (Table 1).

**Table 1: Comparison of different back-end fuel cycles on volume and heat emission of waste**

Concept	Volume (Unit: m <sup>3</sup> /1 MT)	Heat emission after 300-year cooling time (Unit: watts/tonne)
Pyroprocessing	0.37	2.00E+00
Direct disposal	3.06	1.34E+02

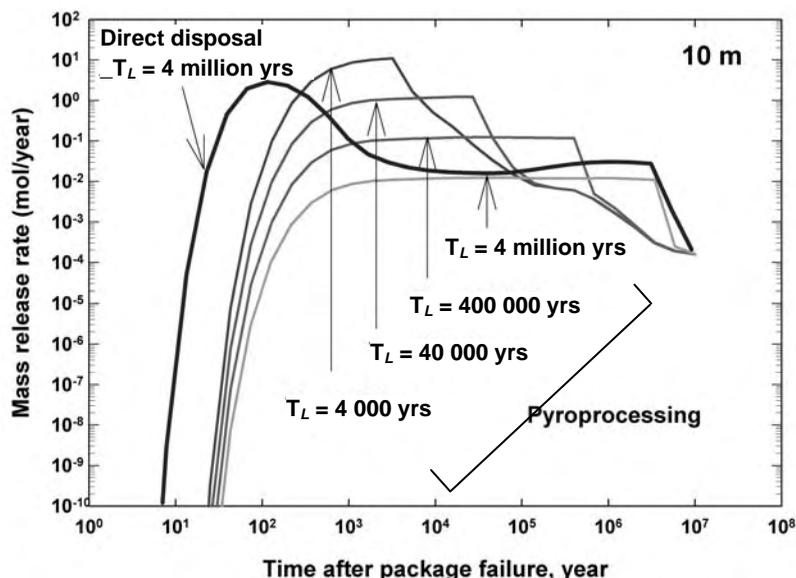
These reduction effects for the volume and heat emission would suggest a potential reduction of the repository footprint from the KRS concept to the A-KRS concept. Actually, KAERI claims that this would be one of the major advantages that would be expected by applying the KIEP-21 and A-KRS concepts [2]. However, we recognise the necessity of performing more careful and comprehensive analysis to finally conclude if such an advantage actually exists. First, the

repository footprint is determined by various site-specific conditions, such as rock-mechanical stability, heat and hydraulic conductivity, and engineered-barrier conditions, such as stability and heat conductivity of the buffer material. Second, we need to take into account the effects of separated uranium and transuranium elements fairly. It is expected that separated TRU will be used as fuel materials in a fast reactor in the KIEP-21 concept. If so, additional ILW and HLW will be generated from such recycle. If they are not recycled, they will be disposed of in a repository eventually, which would not make a difference with the direct disposal case. Separated uranium is often considered as a low-level waste. However, its radioactivity and radiotoxicity increase with time, and eventually reach the same level as the original uranium ore and resulting mill tailings. If it is recycled, then similar considerations need to be made to those for the TRU recycle in terms of repository impacts. Third, there is potentially an inter-generational issue. In the A-KRS concept, a certain length of cooling is included to accommodate ILW and HLW in a smaller footprint. This would be fair while such management cost is paid off by the revenue by nuclear power utilisation. But, when nuclear utilisation has phased out in the future, operation of the interim storage part of the system must be maintained until the pre-set cooling time ends. This could be a potentially significant financial and institutional burden for future generations.

### Waste form durability effect

To observe the effects of waste form durability, we undertook numerical parametric studies by changing the leach time of waste forms for the pyroprocessing case, ranging from 4 000 years to 4 million years (Figure 4). For comparison, the result for the direct disposal case is also included in Figure 4. Because the dominant radionuclides are fission product isotopes, which are released congruently with waste form dissolution, the peak values decrease proportionally as the leach time increases. The duration of time for the plateau corresponds to the leach time.

**Figure 4: Waste form durability effect on total mass release rate of radionuclides at the 10-m location for pyroprocessing wastes**



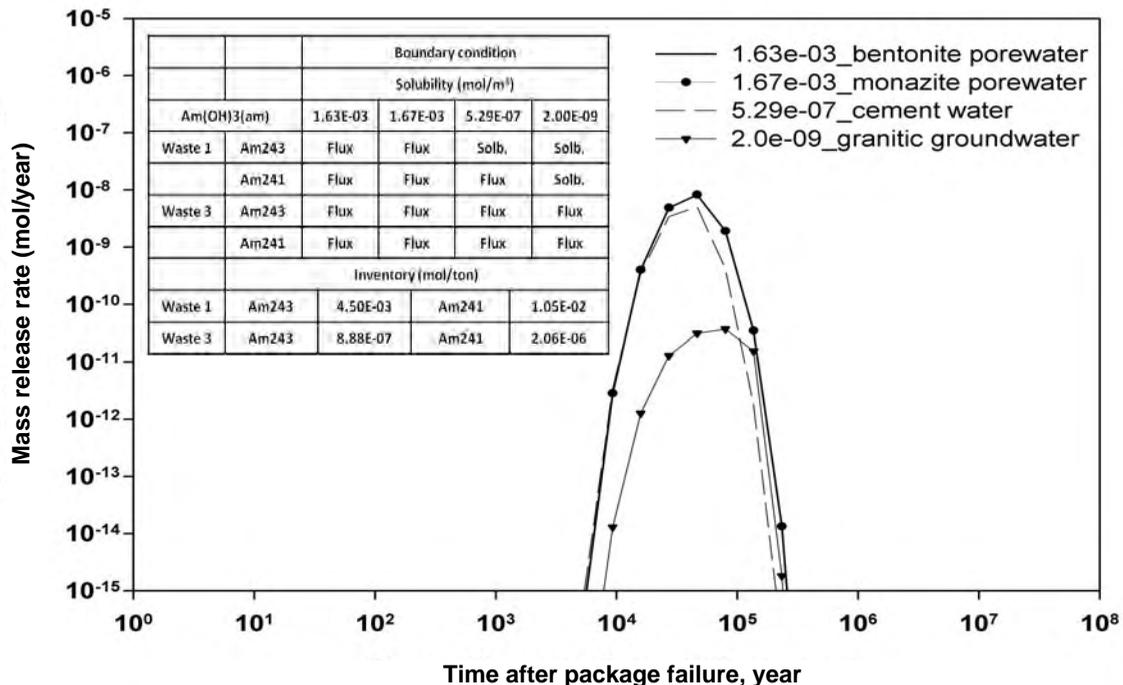
### Solubility effect

To take into account the coupled phenomena among waste forms, EBS materials, groundwater and radionuclides in a system performance assessment, we have evaluated the solubility of radionuclides in surrounding engineered and geological environments. Different waste forms and EBS configurations should affect chemical conditions in the EBS and NBS, resulting in

significant differences in radionuclide solubilities and sorption distribution coefficients. As a case study, solubility of americium (Am) was calculated by using PHREEQC in different geochemical conditions such as granitic groundwater, cement water, bentonite pore water and monazite pore water.

Figure 5 shows the results of total mass release rate of americium with different solubility values. The values of solubility obtained for the pore water equilibrated with bentonite and the pore water equilibrated with monazite are similar. The solubility for the cement water, which is considered to exist in the outer boundary of the engineered barriers or the disposal tunnel surface, was determined to be four orders of magnitude lower. The solubility for the groundwater equilibrated with granite, which is observed in the host rock, is even lower by almost two orders of magnitude than that in the cement water. The table inset in Figure 5 shows the boundary conditions for the four waste forms in the pyroprocessing case under the assumption that these values of solubility are applied at the waste form dissolution location. “Flux” indicates that americium is released congruently with the waste form dissolution with that particular solubility value, while “solb” means that americium concentration at the waste form dissolution location is constant at the solubility value. With the solubility value for granite water, release of americium is limited by its solubility, while with solubility values for bentonite or monazite pore waters, the release is congruent with the waste form dissolution. The solubility value for the cement water indicates the border case below which the solubility limitation is on. It means most americium released from waste form and transported through a bentonite buffer would be precipitated at the outer boundary of EBS in this geochemical system. Further studies on models with high fidelity and reality for the coupled phenomena among waste forms, EBS materials, groundwater and radionuclides are necessary.

Figure 5: Total mass release rate of americium with different solubility values



### Conclusions

In the near field, it has been observed that the total radionuclide release rate could be smaller in the case of A-KRS coupled with pyroprocessing than the case of direct disposal of used PWR fuel in KRS. However, this is not because of separation of TRU from the used PWR fuels, but because

of solidification of  $^{129}\text{I}$  in a dedicated waste form, which exhibits slower release of iodine than from used PWR fuel containing  $^{129}\text{I}$  in the gap between fuel pellets and cladding. TRU elements remain as precipitates in the engineered barrier because of low solubilities. In the far field, therefore, fission products are the main contributors.

Furthermore, differences in TRU inventories and waste form performance do not make a significant difference, due to the assumed mechanism of radionuclide retention and dispersal in the far field. This part of modelling is dependent on conditions and assumptions for the geological formations of the site. Until the site has been determined, this part of the analysis remains stylised, and so comparison of two repository concepts by this release rate at the far field location should be interpreted in a stylised manner.

From the preliminary parametric study, it has been shown that geochemical conditions in the near field region would be significantly different, dependent on material configurations in that region. New waste forms, different radionuclide loadings and modification of the engineered barriers, such as addition of cement, would change the principal geochemical parameters such as radionuclide solubilities and sorption distribution coefficients. Thus, for a more meaningful comparison of two repository concepts, we need to make a realistic assessment of repository performance, for which we need to achieve:

- more detailed heat transfer analysis to determine repository configurations;
- more detailed geochemical analysis to determine waste form dissolution, release of radionuclides from waste forms, and transport of radionuclides in the engineered barrier.

Though reduction of the volume and heat emission is seemingly a major advantage that could be expected by applying the KIEP-21 and A-KRS concepts, more careful and comprehensive analysis is indispensable to derive a final conclusion concerning the system's benefits.

### Acknowledgements

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## **Session III**

### **Transmutation Fuels and Targets**

***Chairs: K. Minato, K. Pasamehmetoglu***



## Minor actinide transmutation in an accelerator-driven system prototype: Results from fuel developments within the European programme EUROTRANS

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### Abstract

*Studies on fuel development within the EUROTRANS project, motivated by assessing industrial practicability for actinide transmutation, have provided a wide range of results.*

*Several cores loaded with primary TRU fuel candidates: MgO-cermer and <sup>enr</sup>Mo-cermet, have been designed and optima meeting the specifications have been found. Preliminary thermo-mechanical calculations of the hottest fuel pins have provided evidence of good performances. Based on current analyses and knowledge, these fuels do not pose safety limits. Nevertheless, safety margins for <sup>enr</sup>Mo-cermet fuel are considerably higher than those for MgO-cermer.*

*Irradiation tests: FUTURIX-FTA and HELIOS, on Mo-cermet and MgO-cermer fuels have been completed. Helium behaviour in inert matrices during irradiation was investigated within the BODEX irradiation test.*

*Thermal properties of a large range of fuels have been assessed. Compatibility tests were performed between: actinide phases, inert matrix candidates, T91 cladding and Pb coolant. Finally, great progress has been made in Pu-Am-O phase diagram investigation.*

*These results reinforce the interest of both MgO-cermer and <sup>enr</sup>Mo-cermet for the 400 MWth EFIT machine with higher safety margins for the cermet fuel.*

## Introduction

The work performed on fuel developments under the EUROTRANS European project umbrella [1] deals with design, development and qualification under representative conditions of promising U-free oxide fuel concepts for the 400 MWth European Facility for Industrial Transmutation. Outcomes consist of recommendations about fuel designs and performances. Hence, fuel performances and safety of the preliminary EFIT core designs have been evaluated. In-pile and out-of-pile experiments have been carried out to gain knowledge about fabrication, properties and behaviour under irradiation of these challenging fuel concepts. The current paper focuses on experimental results gained within the project, after which preliminary fuel selection as well as fuel element design and performance considerations are described.

## Fuel candidates

Emphasis has been placed upon oxide fuels in compliance with the guidance of the European Technical Working Group on ADS [2] and:

- promising results according to performance, safety and fabricability criteria as well as initial experimental feedback on (Pu,Am)O<sub>2</sub> fabrication and out-of-pile characterisations, gained within the FP5 FUTURE programme (2002-2006) [3];
- a strong synergy with R&D programmes on transmutation targets [4];
- a broad industrial experience on oxide fuel fabrication for critical reactors.

Nitride-based fuels have been considered as a possible back-up solution. Indeed, though their high attractiveness has been confirmed by the results of the FP5 CONFIRM programme (2001-2008) [5] and ongoing developments in Japan, knowledge and technical know-how in Europe remains limited. Thus these fuels are considered to be at an early stage of development with longer-term R&D activities still required.

Two primary oxide candidates were selected at the beginning of the project, from a ranking procedure based on criteria of: fabrication, hydro-reprocessing, secondary waste stream, high-temperature stability as well as expected neutronic, transmutation and thermo-mechanical performances and safety margin to failure. These primary candidates are: MgO ceramic-ceramic (cercer) and <sup>enr</sup>Mo ceramic-metallic (cermet) fuels, consisting of (Pu,MA)O<sub>2</sub> particles dispersed respectively in a magnesia matrix or a molybdenum matrix enriched in <sup>3/2</sup>Mo (to decrease Mo neutronic penalties as well as to prevent the production of the long-lived <sup>99</sup>Tc by neutron capture in <sup>98</sup>Mo). Finally, ZrO<sub>2</sub> inert matrix fuels have been considered as a back-up option due to expected difficulties for hydro-reprocessing.

## Fuel element design and performance considerations

The EFIT machine, intended to be operational around 2040, is a pure lead-cooled ADS reactor of 400 MWth dedicated to minor actinide transmutation. The reference working hypotheses for its conceptual design have been as follows [6]: a proton beam of 800 MeV impinges on a windowless lead target providing the neutron source for the subcritical system. The inlet and outlet temperatures of the lead coolant in the ADS core are respectively 400 and 480°C. Material for cladding is a ferritic-martensitic steel (FMS) T91. The assumed plutonium and minor actinide isotopic compositions come from a mixture of 45 GWd/tonne spent UO<sub>2</sub> and MOX fuels in a 90/10 ratio [7].

EFIT core and fuel element designs have been performed with the objective of a transmutation rate of 42 kg MA/TWh<sub>th</sub> with a zero net balance of Pu (hereafter called the “42/0 approach”), as well as assuring a flat power distribution, a low reactivity swing, a low core pressure drop and safety requirements.

Dealing with cermet fuel, from a first three-zone core design, loaded with fuel elements whose characteristics are quite similar to those of the cercer core reference design (see Table 1), improvements have been performed [8, 9] to limit the high reactivity variation swing. These advanced cores designed by lowering the Pu/MA ratio, increasing core fuel fraction with large fuel pellet diameters and reducing the pin number per SA to maintain thermo-hydraulic characteristics have shown that similar performances (see Table 2) are achievable for both MgO-cerker and  $^{92}\text{Mo}$ -cermet fuels.

**Table 1: Preliminary design data for fuel pin and sub-assembly**

	MgO-cerker core [2]			$^{92}\text{Mo}$ -cermet core [8]		
	Inner	Intermediate	Outer	Inner	Intermediate	Outer
Pu/MA (at.%)	45.7/54.3			46/54		
Fuel/matrix	57/43	50/50		35/65	43/57	50/50
Fuel pellet diameter (mm)	7.10		8.00	8.00		
Clad inner diameter (mm)	7.42		8.32	8.32		
Clad width ( $\mu\text{m}$ )	600			600		
Fuel pin pitch (mm)	13.63		13.54	13.54		
Pin number/assembly	169			169		
Row number/assembly	7			7		
Assembly number/zone	42	66	72	42	90	84

**Table 2: Transmutation performances of cerker and cermet cores**

	Cerker [6] reference	Cermet [9]		
		Reference	Advanced	
No. pins per S/A	169	169	91	61
Pu/MA	46/54	46/54	40/60	35/65
Pin diameter	7.52-9.52	9.52	13.00	15.87
Fuel volume (%)	23.33	26.73	29.79	31.24
$K_{\text{eff}}$ :				
– Initial	0.9655	0.9820	0.9724	0.9428
– After 1 098 EFPD	0.9654	0.9593	0.9625	0.9455
$\Delta$ (kg/TWh <sub>th</sub> ):				
– Pu	-42.00	-43.67	-43.92	-43.78
– MA:				
U,Am	-3.84	-5.71	+1.06	+7.95
Cm,Np	-38.16	-37.96	-44.98	-51.73
U,Am	+0.44, -47.77	+0.48, 46.80	+0.49, -54.72	+0.50, -63.40
Cm,Np	+10.54, -1.37	+9.70, -0.86	+10.70, -0.96	+12.27, -0.60

Regarding the thermo-mechanic behaviour of the fuels under irradiation, preliminary calculations have been performed with the codes TRAFIC and MACROS, initially designed to model homogeneous fuels, and extended during the project to handle heterogeneous media. As fission gas behaviour in ADS-type fuels is poorly known, parameters were tuned to fit available gas release results on MgO-cerker transmutation targets [10, 11] before performing simulations of the ADS fuel behaviours under EFIT normal operation conditions. Results from TRAFIC for ADS fuels indicate that the thermo-mechanical performances of cerker and cermet hottest pins are acceptable for normal operation conditions (peak-linear powers around  $200 \text{ W.cm}^{-1}$  and operation time of 1 098 equivalent full power days), with higher temperature margins to failure for cermet fuel [12]. These promising first thermo-mechanical results remain nevertheless quite unreliable due to the lack of experimental knowledge concerning basic mechanisms of fuel behaviour under irradiation; further progress is still required.

Regarding safety, analyses have shown [9, 13] that temperature limits for fuel are not violated in either cerker and cermet cores, although the safety margins for the cermet core design are considerably higher. The safety analyses also reveal that the most limiting conditions would originate from the T91 clad.

## Irradiation tests

Even if the behaviour of TRU fuel under irradiation is quite unknown, results of post-irradiation examination (PIE) on irradiated targets with a few per cent of americium have already emphasised [14] the major roles played by irradiation conditions (including temperature), helium production and material swelling due to structure modifications, amorphisation, helium accumulation,... To obtain further information, three irradiation tests were performed on oxide fuels to investigate:

- irradiation effects in EFIT representative conditions with FUTURIX-FTA test [15] in the PHENIX reactor;
- helium behaviour versus temperature and microstructure with HELIOS test in the HFR reactor [16];
- helium build-up and release mechanisms versus temperature in  $^{10}\text{B}$  (as an Am surrogate) doped fuel matrices with BODEX test [17] in the HFR reactor.

The composition of pellets tested within the FUTURIX-FTA and HELIOS experiments are given in Table 3. FUTURIX-FTA compositions specifically address ADS-type fuels. HELIOS compositions range from ADS-type fuels to transmutation targets, in order to link and thus to benefit from the experience gained within the EFFTRA-T4 and -T4bis tests [18] for ADS fuel (and target) developments. The Am content ranges from 0.2 to 1.9 g.cm<sup>-3</sup> in FUTURIX-FTA samples, whereas it is about 0.7 g.cm<sup>-3</sup> in HELIOS samples. These highly radioactive materials were fabricated at laboratory scale in two steps. Am particles were first synthesised using two processes, an oxalic co-precipitation route for cercer compounds [19], and a combination of external gelation and infiltration methods for cermet and homogeneous compositions [20]. The following steps are based on conventional powder metallurgy and were similar for all compositions except HELIOS cercer fuel, whose porosity was tailored to remain open in order to allow helium to escape.

**Table 3: Fuel compositions for FUTURIX-FTA and HELIOS tests**

<b>FUTURIX-FTA</b>	Pu <sub>0.80</sub> Am <sub>0.20</sub> O <sub>2-x</sub> + 86 vol.% Mo	<b>HELIOS</b>	Am <sub>2</sub> Zr <sub>2</sub> O <sub>7</sub> + 80 vol.% MgO
	Pu <sub>0.23</sub> Am <sub>0.24</sub> Zr <sub>0.53</sub> O <sub>2-x</sub> + 60 vol.% Mo		Zr <sub>0.80</sub> Y <sub>0.13</sub> Am <sub>0.07</sub> O <sub>2-x</sub>
	Pu <sub>0.5</sub> Am <sub>0.5</sub> O <sub>1.88</sub> + 80 vol.% MgO		Pu <sub>0.04</sub> Am <sub>0.07</sub> Zr <sub>0.76</sub> Y <sub>0.13</sub> O <sub>2-x</sub>
	Pu <sub>0.2</sub> Am <sub>0.8</sub> O <sub>1.73</sub> + 75 vol.% MgO		Am <sub>0.22</sub> Zr <sub>0.67</sub> Y <sub>0.11</sub> O <sub>2-x</sub> + 71 vol.% Mo
			Pu <sub>0.80</sub> Am <sub>0.20</sub> O <sub>2-x</sub> + 84 vol.% Mo

The FUTURIX-FTA irradiation test is currently complete with an irradiation time of 235 EFPD and cumulative neutron fluence of respectively  $\sim 1.0 \times 10^{23}$  n.cm<sup>-2</sup> and  $\sim 1.4 \times 10^{23}$  n.cm<sup>-2</sup> for cercer and cermet capsules. HELIOS irradiation occurred between 29 April 2009 and 19 February 2010.

Dealing with the BODEX test, the experiment was designed to investigate He-induced swelling behaviour of inert matrices using  $^{10}\text{B}$  as an Am surrogate. Indeed, under irradiation  $^{10}\text{B}$  can produce a large amount of helium in a short period of time due to the large cross-section for the  $^{10}\text{B}(n,\alpha)^7\text{Li}$  reaction. Thus,  $^{10}\text{B}$ - as well as  $^{11}\text{B}$ -doped pellets and blank samples (without B) were fabricated in order to discriminate the effects related to helium production ( $^{10}\text{B}$ -doped samples) from irradiation (blank samples) and chemical ( $^{11}\text{B}$ -doped samples) effects. The boron compounds mixed with MgO, Mo and yttrium-stabilised ZrO<sub>2</sub> (YSZ) – as a back-up candidate matrix – are Mg<sub>3</sub>B<sub>2</sub>O<sub>6</sub>, Mo<sub>2</sub>B and ZrB<sub>2</sub>, respectively. The boron content is about 1 wt.% in order to obtain a similar level of helium production ( $\sim 0.3$ - $0.4$  mmol) after two HFR irradiation cycles (57 EFPD) as with the HELIOS (or EFFTRA-T4) irradiation test. The density of the BODEX samples are quite similar to those for HELIOS (>90%) except for the MgO pellets whose porosity is high (18-24%) and totally open. The irradiation test was performed at two temperatures:  $\sim 1$  073 K and  $\sim 1$  473 K. The experiment included neutron fluence detectors in all capsules and on-line pressure and temperature monitoring for capsules with MgO and Mo pellets, heated at 1 473 K.

Helium release in gas lines and capsule punctures were compared to the calculated amounts produced in  $^{10}\text{B}$ -doped pellets during the irradiation (see Table 4). Helium release is three to four times lower for Mo compared to  $\text{ZrO}_2$  (and  $\text{MgO}$ ), whereas  $\text{ZrO}_2$  and Mo density as well as open porosity before irradiation are quite similar (respectively  $d > 90\%$  and  $P_o \sim 3\pm 1\%$ ).

**Table 4: Helium release in capsules**

References	$^{10}\text{B}$ burn-up (%)	He produced (mmol)	He release (%)	Measuring method
Mo-800	65.8	0.277	N/A	–
Mo-1 200	62.1	0.261	$9\pm 1$	On-line
MgO-800	70.0	0.323	$32\pm 3$	Puncturing
MgO-1 200	63.0	0.296	$35\pm 3$	On-line
YSZ-800	68.4	0.300	N/A	–
YSZ-1 200	58.6	0.256	$26.7\pm 3$	Puncturing

Visual inspections (see Figure 1) point out that non-doped samples (as well as  $^{11}\text{B}$ -doped samples not shown in the figure) remain intact after irradiation. On the other hand,  $^{10}\text{B}$ -doped Mo samples developed cracks during irradiation at both 800 and 1 200°C, whereas  $^{10}\text{B}$ -doped MgO and YSZ samples did not, even if these latter samples were significantly embrittled by the neutron flux.

Swelling and open porosity changes have been assessed by dimensional measurements and helium pycnometry respectively. Regarding Mo, swelling is clearly linked with  $^{10}\text{B}$  presence and temperature range (see Figure 2):

- Pure and  $^{11}\text{B}$ -doped Mo samples show a global stability under irradiation even if some small changes have been measured on height (decrease) and diameter (increase) of the pellets underlining a slight anisotropy.
- $^{10}\text{B}$ -doped samples are subject to volumetric changes that range from about 2% at 800°C to 9% at 1 200°C.

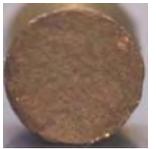
Open porosity changes during irradiation are consistent with visual inspection and swelling results; it closes on pure and  $^{11}\text{B}$ -doped Mo samples, remains constant for  $^{10}\text{B}$ -doped samples which sustained low volumetric changes, and drastically increase for  $^{10}\text{B}$ -doped samples which exhibit both cracks and a significant swelling after irradiation.

For MgO samples (see Figure 3), swelling of  $^{10}\text{B}$ -doped pellets after irradiation at 800°C ( $8\pm 4\%$ ) is larger compared to that after irradiation at 1 200°C ( $3\pm 1.5\%$ ) and a pronounced spread is observed as well. Significant axial and radial deformations have been measured in both irradiation temperature cases. Non  $^{10}\text{B}$ -doped samples show a good stability under irradiation except for the pure MgO sample irradiated at 1 200°C which maybe re-sintered. Open porosity measurements in MgO samples show unimportant changes after irradiation at 1 200°C except for the pure MgO pellet, whose porosity has mostly closed. Regarding  $^{10}\text{B}$ -doped samples irradiated at 800°C, open porosity is significantly higher than that of both pellets before irradiation and high-temperature irradiated pellets.

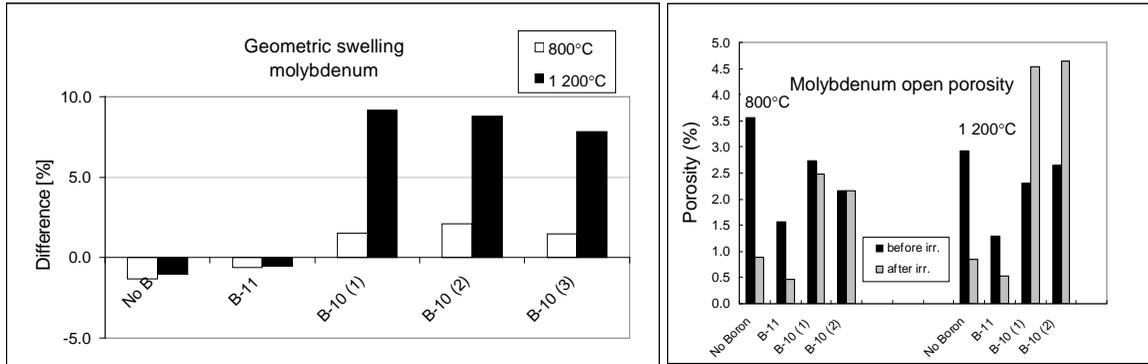
As regards YSZ samples (see Figure 4), all show a limited swelling after irradiation at 800 and 1 200°C, with respectively volumetric average increases for  $^{10}\text{B}$ -doped samples of  $1.2\pm 0.2\%$  and  $4.1\pm 0.7\%$ . Blank samples show a very high stability under irradiation at both temperatures. Open porosity has increased except for the 800°C irradiated blank sample and the 1 200°C irradiated  $^{11}\text{B}$ -doped sample. The increase range is higher in all (concerned) pellets at 800°C.

As a summary, even if microscopy and helium desorption measurements are still ongoing, the BODEX experiment has already provided major results (summarised in Table 5) on helium behaviour in fuel matrix candidates; swelling range is satisfactory in all cases even if open porosity has not been tailored for Mo and YSZ.

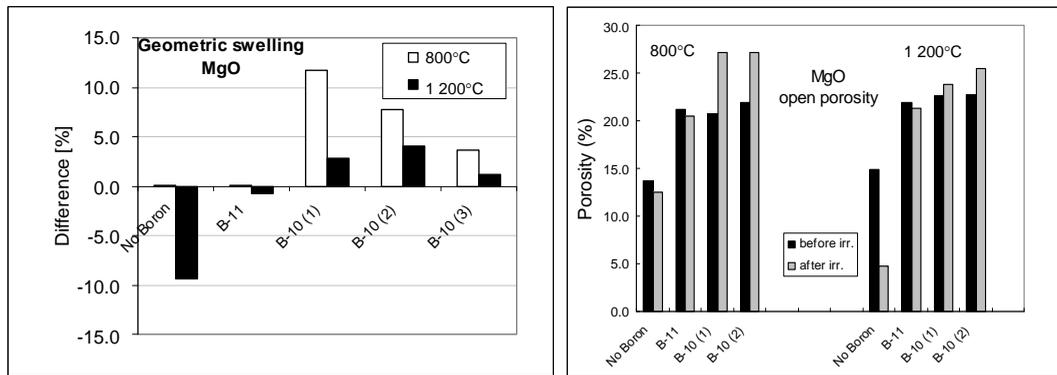
Figure 1: Visual inspection of Mo, MgO and YSZ samples before and after irradiation [17]

		Temperature	Samples without boron		<sup>10</sup> B doped samples	
Mo	800°C		Before irradiation 	After irradiation 	Before irradiation 	After irradiation 
	1 200°C		Before irradiation 	After irradiation 	Before irradiation 	After irradiation 
MgO	1 200°C		Before irradiation 	After irradiation 	Before irradiation 	After irradiation 
YSZ	1 200°C		Before irradiation 	After irradiation 	Before irradiation 	After irradiation 

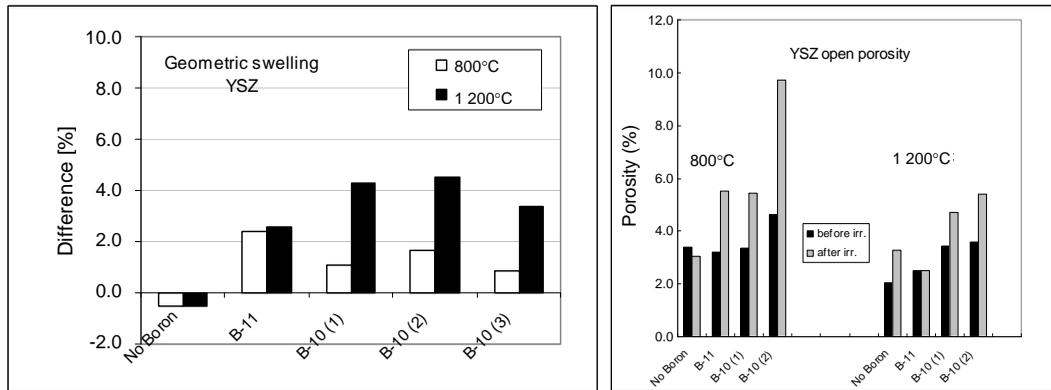
**Figure 2: Molybdenum swelling and open porosity during irradiation [8]**



**Figure 3: Magnesia swelling and open porosity during irradiation [8]**



**Figure 4: Yttrium-stabilised zirconia swelling and open porosity during irradiation [8]**



**Table 5:  $^{10}\text{B}$  matrix irradiation behaviour overview**

Sample reference	$^{10}\text{B}$ burn-up	Open porosity		He release	Swelling (%)	Visual inspection
		BOI	EOI			
Mo-800	65.8%	2.4%	2.3%	N/A	2	Cracks
Mo-1 200	62.1%	2.5%	4.6%	9%	9	
MgO-800	70.0%	21.3%	27.1%	32%	8	Fragile
MgO-1 200	63.0%	22.7%	24.7%	35%	3	
YSZ-800	68.4%	4.0%	7.6%	N/A	1.2	
YSZ-1 200	58.6%	3.5%	5.1%	27%	4.1	

## Out-of-pile measurements

Out-of-pile measurements have aimed at increasing knowledge on the fuel thermal, mechanical and physical-chemical properties in order to improve the databases with relevant and accurate data for the fuel design, fuel performance and safety modelling. Many results have been gained. They are mostly summarised hereafter.

Thermal conductivity experimental measurements made on FUTURIX-FTA fuels up to 2 200 K, have for example pointed out a significant drop at high temperatures ( $T > 1\,500\text{ K}$ ) for cermet fuels, which were not predicted by calculations based on phase mixing models [7].

Vaporisation and melting temperature measurements made on cermet fuels under vacuum and neutral atmosphere, for closed and open systems, have confirmed the higher stability of Mo ( $>2\,300\text{ K}$ ) compared to MgO ( $T \sim 2\,130\text{ K}$ ). Volatilisation species and temperature ranges have also been made clear for MgO and TRU oxides [21]; the MgO vaporisation begins at 1 750-1 800 K, drastically increases between 1 800 and 1 900 K and the major gaseous species is Mg. Regarding TRU gaseous compounds, Am species are detectable at 1 800 K but amounts become significant above 2 200 K; Pu species are recorded above 2 000 K.

Chemical compatibility tests were performed [22] at 1 300 and 1 800 K under different atmospheres (air, Ar, Ar/H<sub>2</sub>) between PuO<sub>2</sub>, AmO<sub>2</sub>, Pu<sub>0.5</sub>Am<sub>0.5</sub>O<sub>2</sub> compounds and inert matrices such as MgO and Mo. The results have not shown any interaction between PuO<sub>2</sub> or Pu<sub>0.5</sub>Am<sub>0.5</sub>O<sub>2</sub> with MgO or Mo under any atmosphere, whereas for tests involving AmO<sub>2</sub> under argon, whatever the inert matrix, undetermined X-ray diffraction peaks are observed.

Chemical compatibility tests performed between T91 clad and monolithic samples of Mo and MgO as well as TRU oxides, under Ar atmosphere during 100 h at 550°C (to simulate inner clad temperature under normal operating conditions), have not pointed out any interaction in T91/Mo and T91/MgO systems [12]. Regarding T91/TRU-oxide systems, slight areas of interaction locally observed on the edge of Pu<sub>x</sub>Am<sub>1-x</sub>O<sub>2</sub> opposite T91 for  $x \geq 0.5$  lead to the preliminary conclusion that the fissile particles have to be surrounded by the inert matrix to prevent interactions with the clad.

No interaction between Pb (EFIT coolant) and Mo or MgO compounds has been detected by differential calorimetry tests up to 1 000°C.

Finally, experimental investigations on the Pu-Am-O phase diagram have provided relevant data to partially build the Pu<sub>2</sub>O<sub>3</sub>-Am<sub>2</sub>O<sub>3</sub> phase diagram [12]. They have further pointed out that Am drives the reduction process in the oxygen sub-stoichiometric area.

## Conclusion

Studies on fuel development within the EUROTRANS project, motivated by assessing the industrial practicability of actinide transmutation, have provided a wide range of results on oxide fuels which have mostly been reviewed in this paper. The interest of both MgO-cermet and <sup>enr</sup>Mo-cermet is reinforced. The ranking between these two primary candidates seems

nevertheless premature without the PIE results of the HELIOS and FUTURIX-FTA fuels, which are under way within the FP-7 FAIRFUELS programme.

Beyond the expected PIE of HELIOS and FUTURIX-FTA fuels, EFIT fuel qualification will require additional developments to demonstrate that the “fuel product fabricated in accordance with a specification behaves as assumed or described in the applicable licensing safety case, and with the reliability necessary for economic operation of the reactor” [23]. Therefore, the next steps regarding ADS fuel R&D should include achievements in the areas of predictive modelling of fuel behaviour under irradiation, fabrication techniques and processes for industrial-scale transposition, experiments to improve the fuel behaviour understanding under off-normal operating conditions, fuel recycling including reprocessability (hydro- and pyroprocessing) and secondary waste management, properties measurements on irradiated fuels, irradiation tests on ADS fuel reference compositions and designs under nominal conditions of flux, temperature and doses.

### Acknowledgements

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## Development of minor actinide transmutation by CRIEPI

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### Abstract

*The Central Research Institute of Electric Power Industry (CRIEPI) has developed minor actinide (MA) transmutation technology using metal fuel fast breeder reactors (FBR) since the 1980s. The metal fuel FBR has the advantage of efficient MA transmutation due to its high neutron energy and high fuel density. The MA generated in light water reactors (LWR) can be consumed in the metal fuel FBR of the same capacity as LWR by loading homogeneously 2 wt.% MA in the reactor core. Considering the possible fuel contamination with rare-earth (RE) fission products, CRIEPI assessed U-Pu-Zr-MA-RE alloys in co-operation with the Joint Research Centre, Institute for Transuranium Elements (ITU). On the basis of the assessment results, the irradiation experiment of U-19Pu-10Zr metal fuel pins containing U-19Pu-10Zr-2MA-2RE, U-19Pu-10Zr-5MA or U-19Pu-10Zr-5MA-5RE (in wt.%) alloy rods was carried out in the fast reactor Phénix, France. At present, the post-irradiation examination is in progress at ITU.*

## Introduction

Recovering the minor actinides (MA) from light water reactor (LWR) spent fuel and utilising them in fast breeder reactors (FBR) is a promising concept to significantly reduce long-term radiotoxicity and heat generation in nuclear waste [1, 2]. The Central Research Institute of Electric Power Industry (CRIEPI) proposes that the MA recovered from the LWR spent fuel or high-level liquid waste be recycled in the metal fuel cycle along with uranium and plutonium [2-7]. The metal fuel cycle that consists of uranium-plutonium-zirconium (U-Pu-Zr) fuel FBR and pyroreprocessing is entirely compatible with MA recycling. The metal fuel FBR have advantageous characteristics for MA transmutation, e.g. a high heavy-metal density and a hard-neutron spectrum [8]. However, MA content in commercial FBR fuel is limited to a few per cent to keep the neutronic safety parameters acceptable [8]. Consequently, the MA content in the FBR fuel required for recycling the MA from LWR is evaluated through mass flow analysis of Pu and MA (= TRU) during the transition period from LWR to FBR [9]. At this stage, the characterisation examinations and irradiation experiment for MA-containing U-Pu-Zr alloys was carried out in co-operation with the Joint Research Centre, Institute for Transuranium Elements (ITU) [3-7].

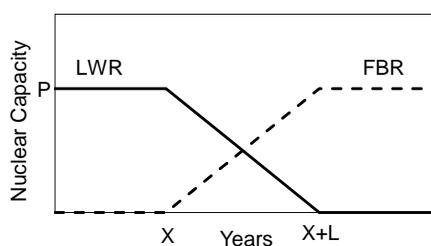
### Mass balance analysis for fuel cycle in the future [9]

The expected MA content in the recycled FBR fuels is evaluated through mass flow analysis of TRU during the transition period from LWR to metal fuel FBR in the future.

#### Transition scenario from LWR to FBR

Figure 1 shows the conceptual transition scheme from LWR to FBR assumed for the evaluation. It is supposed that a fixed scale of power generation ( $P$ -GWe) by LWR is continued for a certain period ( $X$  years) until the FBR starts to be introduced. Assuming that the reactor lifetime is  $L$  years, an average of  $P/L$ -GWe of reactors reaches the lifetime and is replaced by new ones each year. After the period  $X$ , the LWR are replaced by FBR according to the lifetime.

**Figure 1: Conceptual transition scheme from LWR to FBR**



The TRU generated from the LWR are accumulated for  $X$  years and recovered from the spent fuel to be recycled as the start-up fuel for FBR either by conventional reprocessing or pyropartitioning. The remaining LWR continue to generate TRU until  $X + L$  years when all are replaced by FBR. In this study, it is assumed that the metal fuel FBR and the pyroreprocessing plant for their spent fuel are introduced simultaneously, and thus the TRU from the FBR are also recycled and closed to the fuel cycle. The Pu enrichment is determined to satisfy the neutronic and thermal design specifications for commercial-scale FBR. The TRU from the LWR are supplied to make up for the burn-up losses in the FBR.

#### Burn-up performance of TRU in metal fuel cycle

A large-scale PWR with a discharge burn-up of 48G Wd/t is selected to represent the LWR. The burn-up and following decay calculation is conducted with ORIGEN-2 [10]. The spent fuel is reprocessed after five years or longer cooling time and all the TRU are recovered. An overall

recovery ratio of 99.5% is assumed for all the TRU in accordance with the target value of pyropartitioning technology development [2]. The decontamination factor of rare-earth fission products (RE) is ~10, which means that the recovered MA are accompanied by nearly the same amount of RE.

A design of 1 500 MWe metal fuel FBR with a discharge burn-up of 150 GWd/t is used as a model for the commercial-scale FBR [11]. The major specifications of the reactor core are summarised in Table 1. The neutronic calculations are conducted with a diffusion code that can handle the TRU burn-up and decay chains. The spent fuel discharged from the FBR is reprocessed after four years cooling time, and the recovered TRU are returned to core. The overall recovery ratio of all the TRU is 98% and the decontamination factor of the RE is ~10. Table 2 summarises the obtained feed composition and core performance parameters at the equilibrium cycle. In the case of “no MA make-up”, where no MA from LWR are accepted, the contents of MA in the feed become 0.8 wt.%. In the “MA enriched” case, the MA from the LWR are added in the feed to fill up the specified content of 2 wt.% or 5 wt.%. In each case, about 37% or 53% of the MA in the feed are from the LWR, respectively. Since the safety-related parameters such as Doppler constant and coolant coefficient can be degraded due to the harder spectrum and fast fission of the MA as shown in Table 2, the MA content in the feed should be controlled as low as possible from the point of view of core safety.

**Table 1: Design specifications of metal fuel FBR core**

<b>Output [MWe/MWt]</b>	1 500/3 900
<b>Cycle length [years × batches]</b>	1.5 × 4
<b>Coolant temperature [°C, in/out]</b>	355/510
<b>Max. cladding temperature [°C]</b>	650
<b>Max. linear power [W/cm]</b>	500
<b>Discharge burn-up [GWd/t]</b>	150

**Table 2: FBR feed composition and core performance parameters at equilibrium cycle**

	<b>No MA make-up<sup>1</sup></b>	<b>MA enriched<sup>2</sup></b>	
<b>MA content, wt.%</b>	0.8	2.0	5.0
<b>Pu enrichment, wt.%</b>	16.6	16.4	15.8
<b>Make-up MA ratio<sup>3</sup>, %</b>	–	37	53
<b>Doppler const., × 10<sup>-3</sup>Tdk/dT</b>	-2.3	-2.2	-2.1
<b>Coolant coeff., <math>\alpha</math>/°C</b>	0.25	0.27	0.31

<sup>1</sup> No MA from LWR are accepted.

<sup>2</sup> MA from LWR are accepted.

<sup>3</sup> (MA from the LWR)/(MA in the FBR feed).

### **Results of mass flow evaluation**

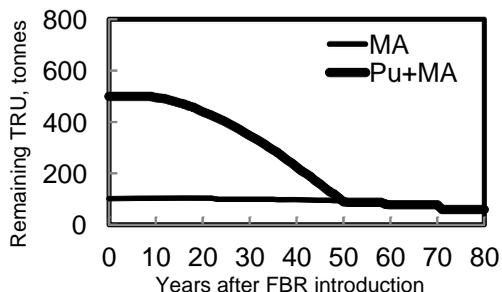
As an example, Figures 2(a), 2(b) and 2(c) show the calculated mass balance of TRU after the 50 GWe of LWR are operated for 50 years. The reactor lifetime is set at 40 years; accordingly 1.25 GWe/year of the LWR are replaced by the metal fuel FBR in which MA content in the fuel is assumed to be 1, 2 or 5 wt.%. As shown in these figures, the introduction of FBR leads to a decrease in the amount of Pu and MA accumulation. However, if the MA content is limited to 1 wt.%, it takes a very long period to consume all MA even after all Pu is consumed in the FBR cycle. On the other hand, in the case of a MA content of 2 wt.%, both Pu and MA are entirely consumed at the almost same time, ~50 years after FBR introduction. If the MA content is increased to 5 wt.%, all MA are consumed in the short term, though Pu accumulation remains for ~50 years. This means that the MA recycling from LWR can be innovated several decades later than FBR introduction. Figure 2(d) shows the mass balance result for the case where the LWR era before FBR introduction is extended to 75 years and the MA content is set to be 2 wt.%. Although

it takes a longer period to consume all TRU in the FBR cycle due to the larger amount of TRU accumulation, the consumption of MA and Pu can be completed almost simultaneously in ~65 years. Therefore, the MA content in the FBR fuel that is sufficient to consume all the Pu and MA from LWR into FBR cycle is estimated to be 2 wt.%. By using a higher MA-content fuel than 2 wt.%, MA can be selectively consumed over a shorter period.

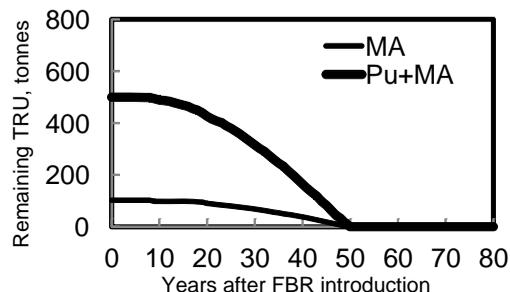
**Figure 2: Analysis results of TRU mass steel and the alloy**

Mass balance during FBR introduction after 50 GWe of LWR are operated for (a), (b), (c) 50 years or (d) 75 years

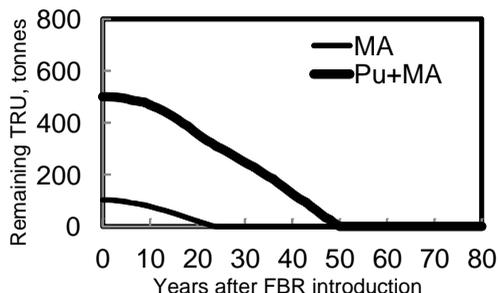
(a) LWR operation = 50 years, MA content = 1 wt.%



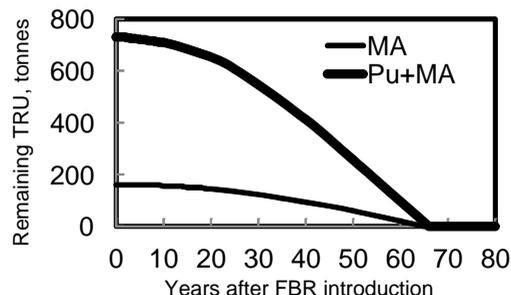
(b) LWR operation = 50 years, MA content = 2 wt.%



(c) LWR operation = 50 years, MA content = 5 wt.%



(d) LWR operation = 75 years, MA content = 2 wt.%



### Characterisation of MA-containing metal fuels

The characteristics of U-Pu-Zr alloy containing MA such as miscibility among the constituents, phase structures, phase transition temperature, melting temperature, density, thermal and mechanical properties, compatibility with stainless steel and solubility into sodium are examined. The MA content is set to be 2 or 5 wt.% based on the mass balance evaluation results, and the RE content is conservatively assumed to be same as MA. The relevant experiments have been performed at ITU.

#### *Miscibility among the constituents of U-Pu-Zr-MA-RE alloys*

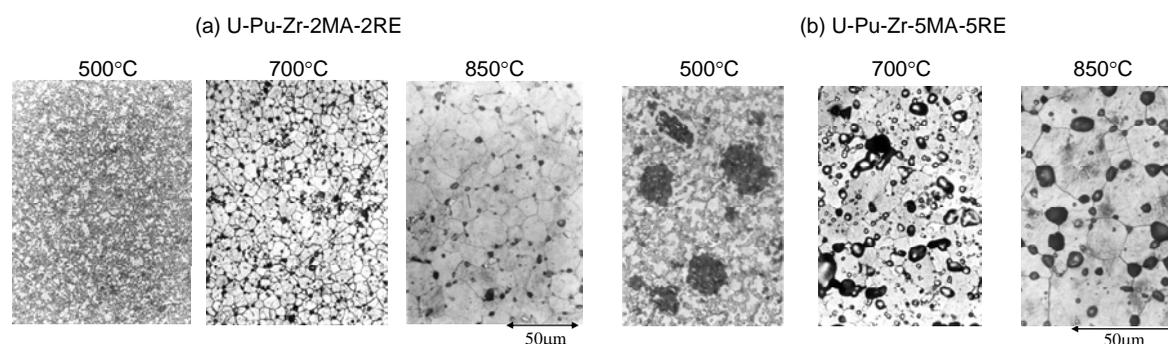
Although U-Pu-Zr-MA alloys without RE were blended homogeneously by a simple arc melting, it was difficult to mix any amount of RE into the U-Pu-Zr-MA alloys because the miscibility between RE and the U-Pu-Zr-MA matrix is low even in the liquid phase. However, if the RE content is reduced to 2% or 5%, the matrix segregation was not found and RE-rich precipitates were uniformly dispersed in the matrix.

#### *Phase structures of annealed U-Pu-Zr-MA-RE alloys*

Two kinds of MA- and RE-containing U-Pu-Zr alloys, U-Pu-Zr-2MA-2RE and U-Pu-Zr-5MA-5RE (wt.%) were annealed at 500, 700 and 850°C for 25-100 hours and quenched rapidly (~1 second) in water to examine phase structures at several temperatures. Figures 3(a) and 3(b) show the

metallography of annealed and etched U-Pu-Zr-2MA-2RE and U-Pu-Zr-5MA-5RE alloy samples, respectively. The compositions of the phases and precipitates observed in U-Pu-Zr-2MA-2RE are summarised in Table 3. The dark-coloured precipitates in the figures were rich in RE and Am, and dispersed almost uniformly in all samples. The sizes of the RE precipitates were  $\sim 3 \mu\text{m}$  in -2MA-2RE and  $\geq 10 \mu\text{m}$  in -5MA-5RE alloy. The matrixes of these alloy samples annealed at  $500^\circ\text{C}$  showed two-phase structure, presumably of  $\zeta + \delta$ . In the alloy samples annealed at  $700^\circ\text{C}$  and  $850^\circ\text{C}$ , the matrixes were single  $\gamma$ -phase and RE-Am precipitates existed at the grain boundary. In Table 3, the Am content in the precipitates of the sample annealed at  $850^\circ\text{C}$  is significantly lower than those of the other samples annealed at lower temperatures. This may be due to Am evaporation during annealing.

**Figure 3: Metallography of annealed and etched U-Pu-Zr alloys containing MA and RE**



**Table 3: Compositions of phases in annealed U-Pu-Zr-2MA-2RE alloy samples [wt.%]**

Anneal conditions	Matrixes							Precipitates						
	U	Np	Pu	Am	Zr	Ce	Nd	U	Np	Pu	Am	Zr	Ce	Nd
500°C, 97 hr	42.9	2.5	22.5	0.9	30.8	0.2	0.2	0.5	0.1	8.1	22.5	0.2	19.6	49.0
	75.6	4.0	17.6	0.2	2.2	0.1	0.2							
700°C, 100 hr	66.8	3.7	19.9	0.4	9.1	0.2	0.1	0.2	0.1	8.1	12.5	0.2	22.6	56.6
850°C, 25 hr	66.2	3.7	20.5	0.4	8.7	0.3	0.1	0.3	0.1	8.1	4.8	0.3	24.8	61.7

### Phase transition temperature of U-Pu-Zr-MA-RE alloys

Phase transition temperatures of U-Pu-Zr-2MA-2RE, U-Pu-Zr-5MA-5RE and U-Pu-Zr alloys were measured using a dilatometry method. The dilatometric curves obtained from  $200^\circ\text{C}$  to  $825^\circ\text{C}$  are shown in Figure 4. These figures indicate two distinctive phase transition temperatures at approximately  $580^\circ\text{C}$  and  $630^\circ\text{C}$  in common among these three samples, and insignificant influence of MA and RE addition up to 5 wt.%. According to U-Pu-Zr ternary phase diagrams [12-14], the U-19Pu-10Zr alloy has phase transitions,  $\zeta + \delta \leftrightarrow \zeta + \gamma$  at  $\sim 600^\circ\text{C}$  and  $\zeta + \gamma \leftrightarrow \gamma$  at  $\sim 650^\circ\text{C}$ , which are in good agreement with the present results.

### Melting temperature

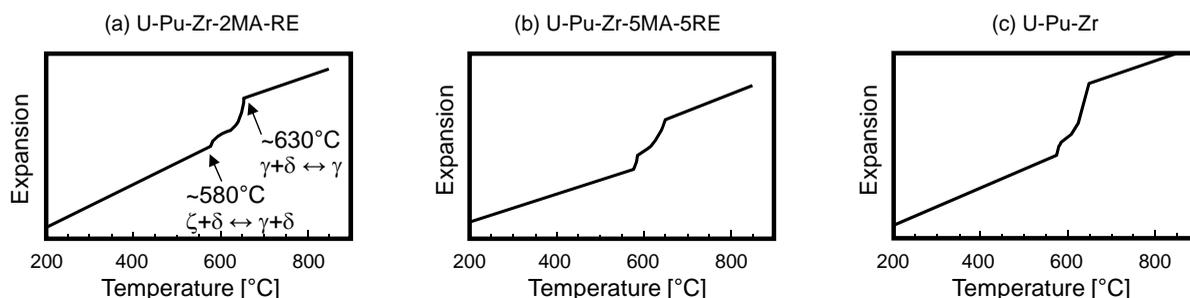
The melting (liquidus) temperatures of U-19Pu-10Zr-5MA-5RE and U-19Pu-10Zr measured by the dilatometry method were  $1207 \pm 10^\circ\text{C}$  and  $1217 \pm 10^\circ\text{C}$ , respectively, which correspond well with previously reported results ( $1214 \pm 75^\circ\text{C}$ ) [15]. The influence of a 5 wt.% MA and RE addition is not significant considering uncertainty in the measurement.

### Density

The measured densities of U-19Pu-10Zr-2MA-2RE, U-19Pu-10Zr-5MA-5RE and U-19Pu-10Zr alloys, which were arc melted and fabricated through the gravity casting method, were 14.73, 14.66 and

15.77 g/cm<sup>3</sup>, respectively. In MA- and RE-containing alloys, the densities were relatively decreased due to the addition of low-density materials as expected. The average density of U-19Pu-10Zr alloys prepared with this method agreed well with the previously reported values (15.8 g/cm<sup>3</sup>) [16].

**Figure 4: Dilatometric curves of U-Pu-Zr alloys containing MA and RE**

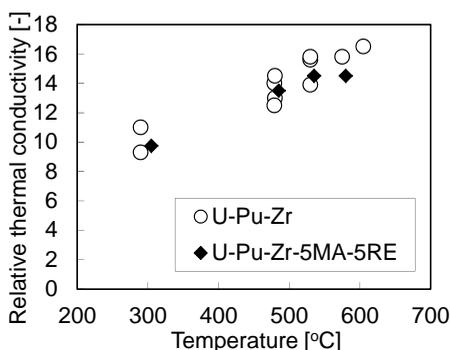


### Thermal conductivity

Thermal conductivities of U-19Pu-10Zr-5MA-5RE and U-19Pu-10Zr alloy samples were measured by a comparative method with pure iron as a reference material. In this study, the measured temperature was limited below 650°C to avoid the plastic deformation of the samples at a high temperature. The measured data, as shown in Figure 5, indicates that the influence of MA and RE addition on thermal conductivity is not significant.

However, further accumulation of the measured data for > 650°C is desirable because a fuel temperature for practical reactor operation is expected to reach to ≥ 850°C.

**Figure 5: Measured thermal conductivity of U-19Pu-10Zr-5MA-5RE and U-19Pu-10Zr alloys**



### Mechanical properties

Table 4 shows the mechanical properties of U-Pu-Zr and U-Pu-Zr-5MA-5RE measured by an ultrasonic method, and suggests that the influence of MA and RE addition on these mechanical properties was not significant.

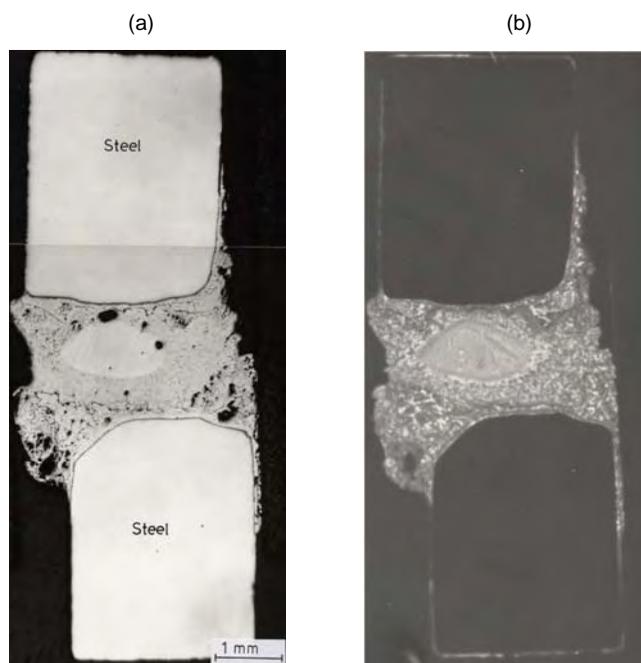
**Table 4: Measured mechanical properties**

	U-Pu-Zr	U-Pu-Zr 5MA-5RE
Sample thickness [mm]	4.30	4.33
Young's modulus [GPa]	93.31	85.22
Shear modulus [GPa]	35.39	32.65
Poisson's ratio	0.32	0.31

### Compatibility with stainless steel [6]

The metallurgical reaction of MA- and RE-containing alloys with stainless steel: Fe-15Ni-15Cr was examined. The diffusion couple of U-Pu-Zr-2MA-2RE/steel or U-Pu-Zr-5MA-5RE/steel was heated in the dilatometer apparatus and the onset temperature of the reaction at the alloy-steel interface was measured. The difference between these two samples was not significant. The compositions of the phases observed in the reaction zones were determined by EPMA. Am-RE-rich precipitates formed in the reaction zone were also rich in Ni. The  $\alpha$ -autoradiography results of the U-Pu-Zr-2MA-2RE/steel couple indicated the density of Am-RE-rich precipitates, which appear brighter in Figure 6(b), and are not high near the contact surface of U-Pu-Zr-2MA-2RE alloy and steel. Am-RE-rich precipitates have no significant effect on the compatibility between fuel alloy and stainless steel cladding.

Figure 6: (a) Optical metallography and (b)  $\alpha$ -autoradiography of the U-Pu-Zr-2MA-2RE/steel couple



### Actinide solubility in sodium

In the metal fuel pins, the fuel alloy comes directly into contact with the liquid Na of the thermal bond material. For that reason, the compatibility of MA-containing alloys with Na was examined in a closed stainless steel capsule. The closed capsule containing U-Pu-Zr-Np-Am-Cm-RE alloy and Na was heated for 1 month at 600°C, and then the solubility of actinides (Pu, Am and Cm) in sodium was analysed by  $\alpha$ -spectrometry. Visual inspections after heating indicated no observable interaction between the stainless steel and the alloy. Table 5 shows the amount of dissolved actinides in Na, and indicates the quite low solubility of these actinides.

Table 5: Dissolution of MA in Na

Actinides	Dissolved amount [ $\mu\text{g}$ ]	Solubility [ $\mu\text{g/g-Na}$ ]
Pu	0.8	0.014
Am	0.36	0.0064
Cm	0.002	0.00004

## Fabrication of MA-containing metal fuels for irradiation experiments

On the basis of the results of characterisation experiments on U-Pu-Zr-MA-RE alloys, three types of MA-containing alloys (U-19Pu-10Zr-2MA-2RE, U-19Pu-10Zr-5MA-5RE and U-19Pu-10Zr-5MA) and a standard ternary alloy (U-19Pu-10Zr) were selected for irradiation experiments in Phénix, France. The arc melted fuel alloys were cast into metal fuel rods in yttria moulds which were compatible with the molten fuel alloys. The cast fuel rods are shown in Figure 7.

**Figure 7: Cast fuel rods**



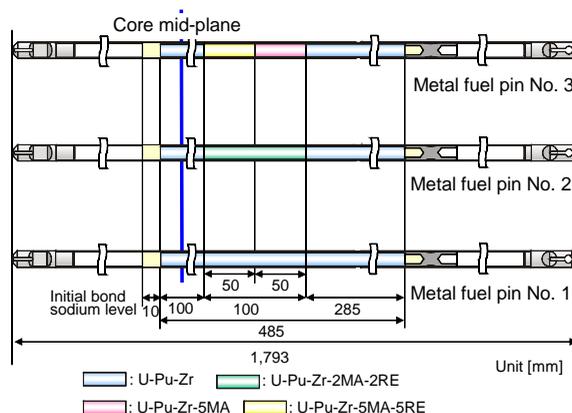
The cladding material is austenitic steel, CW15-15Ti [17, 18]. The cast fuel rods were cut into lengths of 20-50 mm and then inserted in the cladding tubes with metal sodium. The specifications of the fabricated fuel pins are shown in Table 6. The MA-containing alloys were sandwiched by U-19Pu-10Zr rods with 100 mm and 285 mm length, thus the total length of the fuel stack was 485 mm, as shown in Figure 8. For the irradiation experiment, nine metal fuel pins were prepared as follows:

- Pin #1: Three pins of reference U-19Pu-10Zr without MA and RE.
- Pin #2: Three pins including U-19Pu-10Zr-2MA-2RE.
- Pin #3: Three pins including U-19Pu-10Zr-5MA/-5MA-5RE.

**Table 6: Specifications of metal fuel pins for irradiation experiment**

<b>Pin length</b>	1 793 mm
<b>Outer cladding diameter</b>	6.55 mm
<b>Cladding thickness</b>	0.45 mm
<b>Fuel stack length</b>	485 mm
<b>Fuel rod diameter</b>	4.9 mm
<b>Fuel smear density</b>	75.2% TD
<b>Plenum length</b>	464 mm

**Figure 8: Schematic views of test fuel pins**



## Irradiation experiment and post-irradiation examinations [19-21]

Three types of metal fuel pins of different composition, described in the previous section, were arranged in an irradiation capsule, along with 16 oxide fuel pins. Three irradiation capsules of identical configuration were prepared to achieve different burn-ups of ~2.5 at.% (METAPHIX-1), ~7 at.% (METAPHIX-2) and ~10 at.% (METAPHIX-3).

The irradiation experiments of METAPHIX-1, -2 and -3 were successfully completed by May 2008. After cooling time, it was confirmed through non-destructive tests (NDT) that no significant damage on the fuel pins occurred during the irradiation experiments up to the high burn-up of ~10 at.%.

For the METAPHIX-1 and -2 pins, which have already been transported to ITU, the detailed destructive post-irradiation examinations (PIE) are presently under way. The PIE results of gamma-ray spectrometry and plenum gas analysis suggest that up to ~7 at.% burn-up, fuel swelling and fission gas release of irradiated fuel pins including U-Pu-Zr fuel alloys containing  $\leq 5$  wt.% MA and  $\leq 5$ wt.% RE are essentially the same as those of MA- and RE-free U-Pu-Zr fuel pins.

### Optical metallography of ~2.5 at.% burn-up fuel alloys

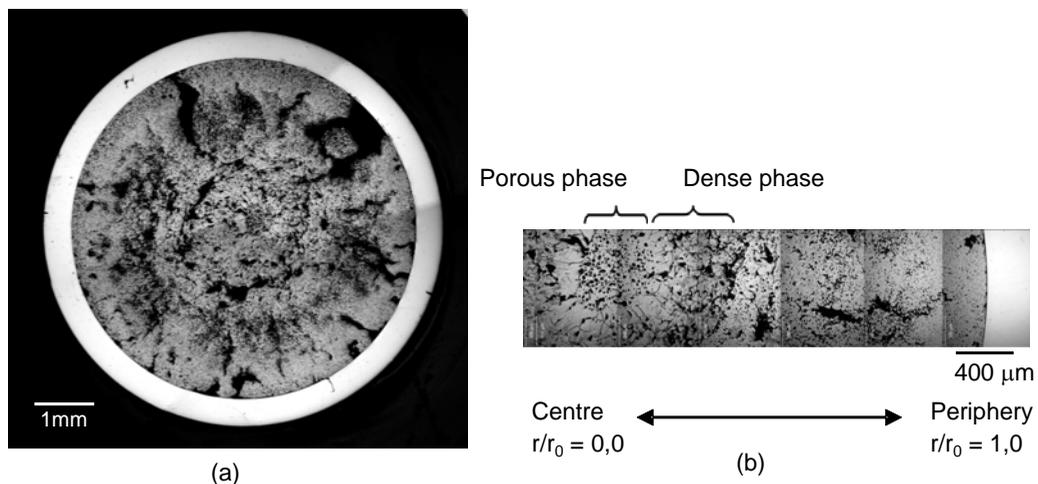
Cross-sectional samples of the MA-containing metal fuels irradiated up to ~2.5 at.% burn-up (METAPHIX-1) were prepared for metallographic examinations, as were the standard samples of U-19Pu-10Zr ternary fuels. The representative observation results are discussed as follows.

#### Sample #1: U-19Pu-10Zr

Figure 9(a) shows the cross-sectional metallography of Sample #1: U-19Pu-10Zr alloy irradiated at 405 mm from the bottom of the fuel stack. In this figure, three distinct concentric zones are observed. Figure 9(b) shows the distribution of the fuel morphology from the centre to the periphery. At the centre, a high-temperature porous  $\gamma$ -phase is observed, surrounded by a relatively dense ( $\gamma + \zeta$ )-phase. The initial gap between fuel and cladding is almost closed.

**Figure 9: Optical metallography of U-19Pu-10Zr fuel irradiated at 405 mm from the bottom of the fuel stack**

(a) Cross-sectional overview and (b) fuel morphology along the radius



**Sample #2: U-19Pu-10Zr-2MA-2RE**

Figure 10 shows the cross-sectional metallography of Sample #2: U-19Pu-10Zr-2MA-2RE alloy irradiated at 370 mm from the bottom of the fuel stack. In this sample, the high-temperature  $\gamma$ -phase does not appear, but a high-density phase similar to ( $\gamma + \zeta$ )-phase in Sample #1 spreads widely from the fuel centre. Specifically, it is deduced that the irradiation temperature of this sample was lower than 630°C. In this sample, moreover, some dark, narrow, layered phases are observed along the grain boundaries, as shown in Figure 10(c). According to the metallography images of the non-irradiated samples containing MA and RE [Figures 3(a) and 3(b)], many dark spots of RE precipitates including MA are uniformly dispersed throughout the alloy. Therefore, it is expected that such precipitates would be redistributed along the grain boundaries in this region during irradiation. As shown in Figure 10(d), some dark spots, inferred to be RE precipitates including MA, are dispersed at the outward region from the middle of this sample ( $r/r_0 \sim 0.3-1.0$ ).

**Figure 10: Optical metallography of U-19Pu-10Zr-2MA-2RE fuel irradiated at 370 mm from the bottom of the fuel stack**

(a) Cross-sectional overview, (b) fuel morphology along the radius and magnified images at (c) central zone and (d) intermediate zone

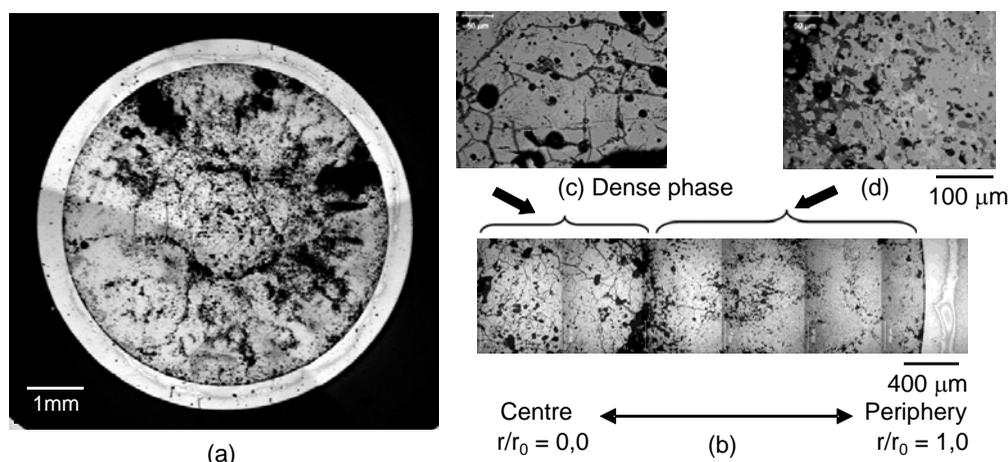
**Sample #3: U-19Pu-10Zr-5MA-5RE**

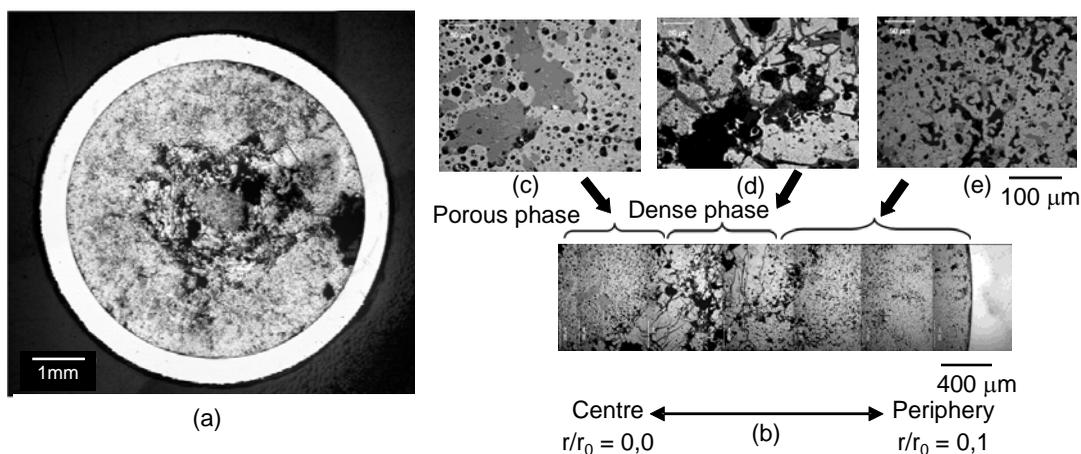
Figure 11 shows the metallography of Sample #3: U-19Pu-10Zr-5MA-5RE alloy irradiated at 370 mm from the bottom of the fuel stack. The matrix phase distribution of Sample #3 is similar to that of Sample #1. On the other hand, some large deposits of more than 100  $\mu\text{m}$  in size appear in the central high-temperature region, as shown in Figure 11(c), and the dark narrow layers similar to those in Samples #2 are observed along the grain boundaries in the surrounding dense region, as shown in Figure 11(d). From the intermediate to the outer region ( $r/r_0 \sim 0.5-1.0$ ), a dispersion of dark spots is clearly seen [Figure 11(e)].

From these metallographic examinations, the characteristics of the irradiated MA- and RE-containing metal fuels are summarised as follows:

- The matrix phase distribution is similar to that of conventional ternary fuels.
- Some large deposits, which are expected to be precipitates of MA and RE, appear in the high-temperature  $\gamma$ -phase.
- In the  $\gamma + \zeta$  mixture phase zone, some dark, narrow, layered phases, which are expected to be precipitates of MA and RE, spread along grain boundaries.
- In low-temperature regions, MA and RE precipitates are dispersed.

**Figure 11: Optical metallography of U-19Pu-10Zr-5MA-5RE fuel irradiated at 370 mm from the bottom of the fuel stack**

(a) Cross-sectional overview, (b) fuel morphology along the radius and magnified images at (c) central zone, (d) intermediate zone and (e) outer zone



## Summary

Mass flow analysis results of Pu and MA during the future transition period from LWR to FBR demonstrate that the MA content in the FBR fuel that is sufficient to consume all the Pu and MA from LWR into the FBR cycle was estimated to be 2 wt.%. Furthermore, by using a higher MA content fuel than 2 wt.%, MA can be selectively consumed over a shorter-term period.

The characterisation examinations of U-Pu-Zr alloys containing MA and RE revealed that Am-RE-rich precipitates are dispersed almost uniformly in the alloy provided that the individual amounts of MA and RE added are limited to 2 or 5 wt.%. The basic properties such as elasticity and phase transition temperature of U-Pu-Zr alloys are practically unchanged after the addition of  $\leq 5$  wt.% MA and  $\leq 5$  wt.% RE. Subsequently, U-19Pu-10Zr fuel stacks, including segments of U-19Pu-10Zr-2MA-2RE or U-19Pu-10Zr-5MA/-5MA-5RE alloy rods were fabricated for the irradiation experiment. The irradiated metal fuel pins were discharged from the reactor at  $\sim 2.5$  at.%,  $\sim 7$  at.% and  $\sim 10$  at.% burn-ups. The NDT of the irradiated fuel pins showed that no damage had occurred during the irradiation experiment.

The optical metallographic observations showed the characteristics of the MA- and RE-containing metal fuels irradiated up to  $\sim 2.5$  at.% as follows:

- The fuel morphology is similar to that of conventional ternary fuels.
- Some large deposits appear in the high-temperature  $\gamma$ -phase.
- In the  $\gamma + \zeta$  mixture phase region, some dark, layered phases spread along the grain boundaries.
- In low-temperature regions, MA and RE precipitates are dispersed.

Quantitative examinations on the redistribution behaviour of the fuel constituents or precipitates and MA transmutation performance are presently being conducted.

## Acknowledgements

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## Development programme on minor-actinide-bearing blankets at CEA

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### Abstract

*An experimental programme will be necessary to qualify the recent minor-actinide-bearing blanket (MABB) concept which involves the transmutation of minor actinides on a fertile UO<sub>2</sub> support in a sodium-cooled fast reactor (SFR) with a heterogeneous recycling mode. This paper describes a development programme to study the behaviour of minor-actinide-bearing blankets under irradiation with the goal of qualifying the concept on a fuel sub-assembly scale according to a schedule that is compatible with the deployment of Generation IV SFR.*

*This development programme can be divided into four main phases: fuel selection, feasibility, optimisation and qualification. An irradiation programme is drawn up for each of these phases, taking into account the manufacturing and irradiation means available at each specific period.*

## Introduction

The CEA has recently undertaken a new R&D programme dedicated to the development of the minor-actinide-bearing blanket (MABB) concept. It is currently considered as one of the most promising options for the heterogeneous recycling of minor actinides (MA) in future Generation IV SFR.

The MABB concept is based on (U,MA)O<sub>2</sub> pellets incorporating a large amount of MA (up to 20 wt.%) which are irradiated for a long time (4 100 EFPD) in radial blankets on the periphery of the outer core [1]. The MABB sub-assemblies are rotated 180° at mid-life so as to balance the burn-up of the various pins in each sub-assembly.

Relative to heterogeneous recycling, the MABB option has little impact on the reactor operating parameters and core safety. It is also entirely compatible with the PUREX-type dissolution process for reprocessing, a feature which is not always prevalent in inert matrices. By positioning the MABB sub-assemblies on the outer core periphery (strong radial variations in the neutron flux inside the sub-assemblies) and by rotating these sub-assemblies 180° at mid-life, the material is subjected to very different operating conditions in the sub-assembly. As a consequence, fuel pellet temperatures can range from 500 to 1 500°C during irradiation.

Due to the large production of helium mainly resulting from the <sup>241</sup>Am transmutation chain, these specific operating conditions are likely to cause significant swelling in the MABB pellets. Furthermore, little information is available on (U,MA)O<sub>2</sub> seeing that (U,Am,Np)O<sub>2</sub> targets were irradiated at higher temperatures (1 500-1 900°C) in the SUPERFACT 1 experiment in the Phénix reactor.

Consequently, it has been decided to launch the first two MABB analytical irradiations in the material test reactor (MTR) in order to study gas releases and MABB swelling as a function of the temperature. The first irradiation experiment – MARIOS – is planned in the high flux reactor (HFR) in Petten within the framework of the FAIRFUELS project. It will investigate temperatures ranging between 1 000 and 1 200°C. The second experiment – DIAMINO – will be performed in the OSIRIS reactor (CEA Saclay Centre) and will focus on irradiation at 600 and 800°C. In parallel, the physical properties of non-irradiated MABB will be measured.

The next step will then involve conducting semi-prototypic irradiation tests on sealed rodlets containing a few representative MABB pellets. The latter experiments should be performed in MTR, e.g. OSIRIS, using high-flux screens to harden the neutron spectrum and to get closer to SFR conditions. Prototypic experiments on representative designs must then be performed under steady-state and transient conditions in a fast reactor, i.e. in the ASTRID prototype reactor in France. The future MA-bearing fuel fabrication facility, ALFA [2], at the CEA Marcoule Centre will be used to manufacture such pins. Finally, the last phase will address the qualification of a whole MABB sub-assembly, with a possible intermediate experimental phase on a bundle of pins using an experimental irradiation capsule.

## MABB concept – Current state of knowledge

The characteristics of the fuel element and the MABB sub-assembly have yet to be finalised.

The MABB fuel pin is still in the preliminary design stage for the core of future SFR (V2B version). Its performance cannot be confirmed due to the lack of reliable data on MABB behaviour under irradiation [3]. There is little knowledge on the swelling of the fuel material under irradiation due to the high helium production and the release rates of fission gases and helium from MABB.

This makes it difficult to reliably assess the transmutation capacity and thus the technical feasibility of such a concept. The type of cladding material also has an impact on performance, though it is currently not realistically possible to choose the most suitable steel grade for the concept.

MABB sub-assemblies are characterised by:

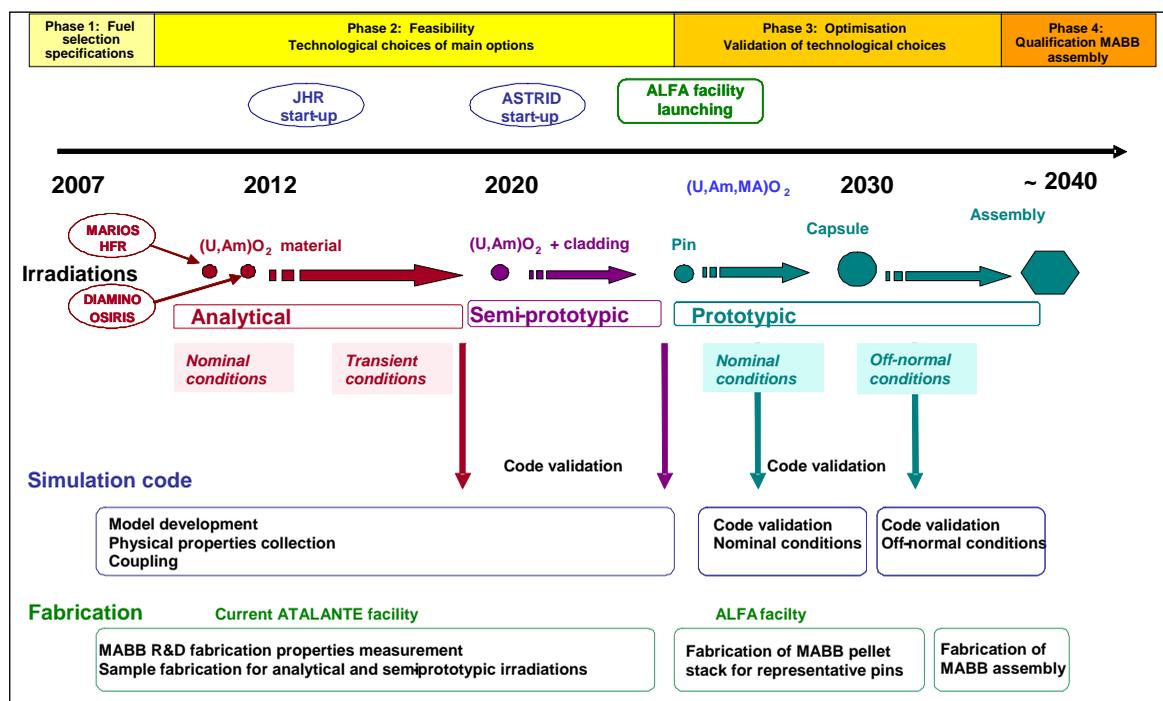
- Long irradiation times currently defined at 4 100 EFPD (10 cycles each lasting 410 EFPD, i.e. twice that of standard fuel), though limited by the behaviour of MABB (internal pressure or pellet-cladding gap, see next point).
- High production of helium due to the transmutation of  $^{241}\text{Am}$  even though the target's thermal conditions are relatively low (operating temperature of 500-1 500°C). This points to significant helium swelling which will have to be quantified and whose activation mechanisms will need to be understood.
- Sub-assembly rotation of 180° at mid-life to homogenise the cladding damage and the transmutation performance of the pins in the sub-assembly. Like the temperature ramp transients, this rotation will be considered as a normal operating transient which will have to be treated carefully when defining the experimental irradiation programme. High gas retention (helium + fission gases) in the fuel can result in additional gas swelling during the transient temperature rise, which will have to be quantified and modelled.
- Pre- and post-irradiation radiological characteristics (neutron emissions and thermal power) outside the qualification range of standard MA-free fuels whose consequences need to be assessed in terms of manufacturing, transport and handling.

## Outline of development programme

A schematic diagram of the development programme for MABB pins and sub-assemblies is given in Figure 1. This programme aims at describing the main phases to be completed prior to qualifying at least one MABB sub-assembly in the ASTRID demonstrator before 2040 – a prerequisite for MABB deployment in industrial-scale reactors.

The different phases requiring the implementation of new manufacturing and irradiation means throughout the project are identified in this programme. It defines a rational irradiation programme with respect to the available means.

Figure 1: Outline of MABB development programme



In the same way as for other nuclear fuel programmes [4], the development programme has been divided into four successive phases: fuel selection, feasibility, optimisation and qualification. The progress of these phases will depend of the material management choices and orientations for future reactor technologies.

This paper describes the four phases and provides an overview of the analytical irradiation experiments to be conducted in the material test reactor (MTR) as part of the second phase.

## Phase 1 – Fuel selection

This phase started in 2007 and aims at compiling information for the MABB pin and sub-assembly specifications. This includes their operating conditions in future SFR and various requirements related to front-end (manufacturing, transport and handling) and back-end (handling, transport and treatment) aspects. These specifications will be used to elaborate the preliminary design studies based on which the out-of-pile and in-pile programmes will be defined in order to qualify the recommended concepts. All available data that can help better understand and model the behaviour of MABB throughout their lifespan will also be analysed during this phase.

The SUPERFACT 1 irradiation experiment conducted in Phénix [5-7] is also a useful source of experimental feedback though it lacks data on the behaviour of this type of material under irradiation. Two pins with  $(U_{0.6}, Am_{0.2}, Np_{0.2})O_{2-x}$  fuel pellets were irradiated for 382 EFPD in the inner core at a relatively high linear power density which is not representative of MABB conditions: about 170 W/cm at the beginning and 270 W/cm at the end. Thermal calculations for this experiment show that the central pellet temperatures would have exceeded 1 500°C during the irradiation, which falls outside the nominal operating range expected for MABB pellets in an SFR blanket ( $\sim 500 < T < 1\ 500^\circ\text{C}$ ). These high thermal conditions in the SUPERFACT 1 pellets most likely led to the full release of the helium produced during irradiation, which will probably not be the case at lower irradiation temperatures.

## Phase 2 – Feasibility

### *Objective and description of Phase 2*

The goal of this phase is to demonstrate the feasibility of the MABB pin concept, i.e. to achieve the predicted transmutation performance while ensuring safe and reliable operation under all circumstances. At the end of this phase, the technological choices for the MABB fuel element concept will be consolidated and the reference manufacturing process for the MABB fuel will be defined. A new version of the code used to simulate the MABB fuel element behaviour under normal operating conditions will also be available.

The chronological order of this programme is:

- A first series of analytical experiments (separate-effect tests) on several fuel samples in available experimental reactors such as OSIRIS, HFR or JHR (soon to be commissioned). These experiments aim at studying the behaviour of the MA-bearing fuel material under irradiation (helium swelling, gas releases) within the temperature range covering all normal conditions in MABB sub-assembly pins. Furthermore, these experiments will set out to correlate the irradiation behaviour with the MABB manufacturing process so as to optimise the microstructure of future MABB fuel pellets for improved performance.
- A second series of “semi-integral” experiments with the fuel and cladding; the geometry will not necessarily be representative of the final fuel pins. These experiments will help better understand the phenomena preventing high transmutation rates, e.g. mechanical and/or chemical interactions between the cladding and the MABB, neutron damage to the cladding, internal pin pressurisation, gas swelling during normal or incident transients, etc. The partial or total representativeness of the future Gen IV SFR technology irradiation conditions is important as the phenomena coming into play depend on the fuel thermochemical conditions.

As the main MA contributor to helium production under irradiation is  $^{241}\text{Am}$ , only this element will be used in the first irradiation experiments (up to Phase 3 of the development programme). The other elements, Np and Cm, will have little impact on the material's behaviour under irradiation, particularly on swelling.

Manufacturing the whole MABB fuel pin for the first integral experiment is a particularly important milestone in the qualification process. This milestone is currently scheduled for sometime around 2025 after the ALFA fabrication facility has been commissioned in Atalante.

The detailed pin specifications resulting from the MABB sub-assembly design studies and a proven, reproducible manufacturing process should both be available by this time. To assess the thermomechanical behaviour of the pins (needed to design the sub-assemblies), a reliable modelling tool should have been developed and validated by 2025 on the basis of feedback from the analytical and semi-integral irradiation experiments. The code reliability will be directly conditioned by the amount of knowledge collected on MABB behaviour during the irradiation experiments.

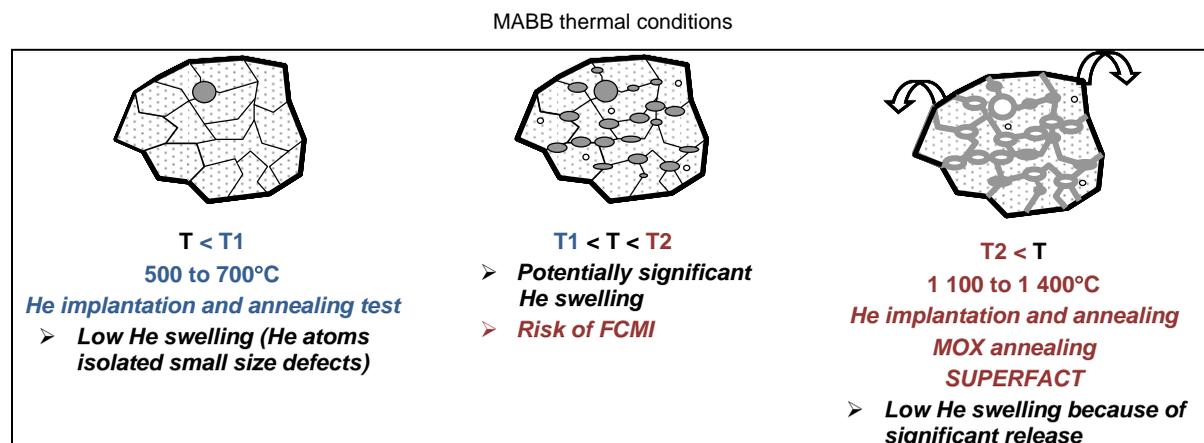
In the long term, studies on the manufacture, reprocessing and handling of MABB fuel pins should also make it possible to validate the technical specifications and determine any uncertainties on specific parameters.

### **Analytical irradiation experiments under nominal operating conditions**

Design of the first analytical experiments began as early as 2007. Two experiments in a thermal reactor are currently being prepared to assess MABB behaviour during normal operation within the MABB operating temperature ranges which were not explored in the SUPERFACT 1 irradiation experiment (1 500°C).

As shown in Figure 2, significant MABB pellet swelling may occur due to helium bubble coalescence in the grain boundaries between the 500-700 (T1) to 1 100-1 400°C (T2) temperature range. It should be pointed out that temperature levels T1 and T2 are likely to change in relation to the burn-up rate.

**Figure 2: Possible behaviour of MABB material as a function of the irradiation temperature**



This swelling phenomenon will be studied and quantified during the MARIOS analytical experiments in the HFR in Petten, as well as during the DIAMINO experiments in the OSIRIS reactor in Saclay. The main objective of these experiments is to control and homogenise the fuel's thermal conditions so as to correlate helium swelling with the fuel temperature. This is why small, thin ( $\text{U}, ^{241}\text{Am}$ ) $\text{O}_{2-x}$  fuel discs will be used in these experiments rather than pellets.

Two types of microstructures will be assessed during these experiments. There will be a "standard" microstructure with a high density (~92 T.D.) achieved with a manufacturing process

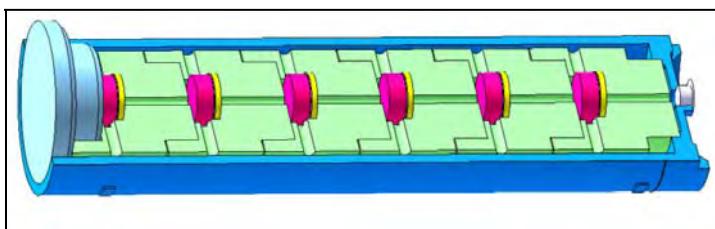
comparable to that used for SFR (U,Pu)O<sub>2</sub> fuels. The other “optimised” microstructure will have a lower density with interconnected open porosity stable under irradiation which should promote the release of helium into the pin free volumes to thus limit swelling.

The two analytical experiments, MARIOS and DIAMINO, are major milestones in the MABB qualification procedure.

#### *MARIOS experiment in the HFR and DIAMINO in the OSIRIS reactor*

The design principle applicable to the MARIOS experiment in the HFR and the DIAMINO experiment in the OSIRIS reactor is illustrated in Figure 3.

**Figure 3: Schematic diagram of a mini-pin with 6 MABB discs**



Small MABB discs ( $\varnothing \approx 5$  mm,  $h = 1.5$  mm) will be placed in molybdenum alloy crucibles. Molybdenum will provide a source of heat by gamma heating and will make it possible to axially remove the heat produced by the sample during irradiation (fission and gamma heating) so as to limit the thermal gradients of the latter. Top axial and radial gaps will be incorporated to allow for free swelling of the sample. An insulating ceramic disc will be added under the MABB sample to balance thermal resistance created by the top axial gap. This measure will limit the thermal gradients inside the MABB discs.

Several mini-pins will be placed inside an irradiation capsule which will then be inserted into the core for irradiation (4 mini-pins for MARIOS and 6 for DIAMINO). Each mini-pin will be used to study an experimental configuration, e.g. 1 target temperature  $\times$  1 microstructure  $\times$  1 helium production rate for DIAMINO. The conditions tested for each experiment are listed in Table 1 below.

**Table 1: Experimental grid for MARIOS and DIAMINO analytical experiments**

Composition		Targeted temperature (°C)			
		600	800	1 000	1 200
MABB (15% Am)	Standard microstructure	DIAMINO	DIAMINO	MARIOS	MARIOS
	Optimised microstructure	DIAMINO	DIAMINO	MARIOS	MARIOS
MABB (7.5% Am)	Standard microstructure		DIAMINO	← Low He production rate	
	Optimised microstructure		DIAMINO		

Two irradiation temperatures ( $\sim 1\,000^\circ\text{C}$  and  $1\,200^\circ\text{C}$ ) will be defined for the two microstructures in the MARIOS experiment. The target neutron fluence must be able to generate a quantity of helium similar to that which will be produced in MABB containing 20% minor actinides loaded in an SFR. This leads to an irradiation time of 310 EFPD for a MABB loaded in position G7 (or a very similar G3 position as back-up) of the HFR core (compared with 4 100 EFPD planned for future SFR). The irradiation is expected to start early 2011 so that the preliminary results on MABB behaviour will be available early 2012.

The DIAMINO experiment will also test two irradiation temperatures (600 and  $800^\circ\text{C}$ ) on the two types of microstructures so as to supplement and readjust results in relation to the MARIOS experiment. The impact of the helium production kinetics will also be studied at  $800^\circ\text{C}$  by irradiating samples with different compositions so as to produce the same quantity of helium

over a period lasting twice as long, i.e. about 400 instead of 200 EFPD. These conditions will be used to check if the irradiation tests under a thermal flux (which notably accelerates the helium production rate) are well suited to MABB when studying the problem of helium swelling. The irradiation device will be equipped with several insulating gas-filling bonds whose composition will be adjusted during irradiation. It will also be equipped with a heater element to precisely adjust the temperature of the MABB discs during the irradiation experiment.

The DIAMINO experiment in the OSIRIS reactor will be conducted about a year later in relation to the MARIOS experiment; irradiation on the first mini-pins should start in mid-2012 to finish in mid-2013.

#### *Manufacturing MABB discs for the MARIOS experiment*

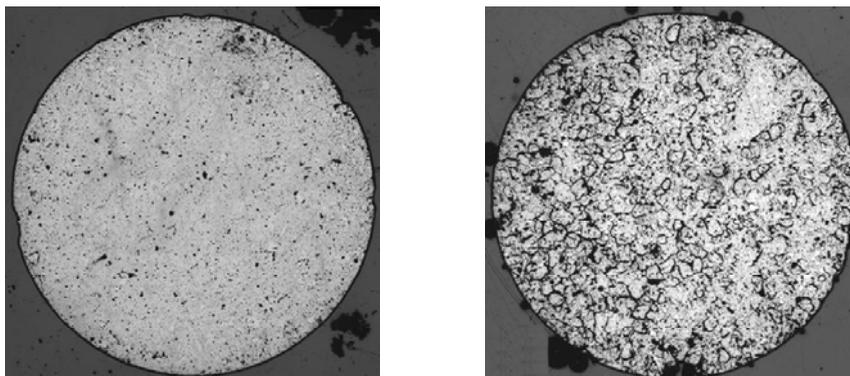
The (U,Am)O<sub>2</sub> discs for the mini-pins in the MARIOS experiment were manufactured with a powder metallurgy process in the Atalante facility in Marcoule and then delivered to Petten in the first half of 2010. As mentioned above, two different microstructures were manufactured:

- a standard microstructure with a high density (~92% TD);
- an optimised microstructure with open, interconnected porosity (~88% TD).

A major R&D phase was required to develop the manufacturing processes, especially to elaborate a pore-forming material for optimised microstructures capable of resisting radiolysis and thermolysis phenomena owing to the high americium content.

The microstructures developed during this R&D phase are shown below in Figure 4.

**Figure 4: Macrograph of standard and optimised MABB pellets ( $\varnothing \approx 4.5$  mm)**



The manufacturing process involved the successive dosing and mixing of UO<sub>2</sub> and AmO<sub>2</sub> powders, followed by dry grinding, press shaping and sintering at 1750°C in a reducing atmosphere. The pore-forming material for the optimised microstructure was added to the powder mixture before pressing. X-ray diffraction analysis showed that the material had a single fluorite-type phase after sintering for both microstructures.

The sintered pellets were then cut and polished to make the 1.5 mm thick discs.

The physical characteristics of several MABB compositions (different Am contents and O/M ratios) will soon be measured in view of optimising the design of the fuel pins to undergo reactor irradiation in the future. The main characteristics to be measured are: thermal conductivity, thermal expansion, melting point and vapour pressure.

#### ***Semi-integral irradiation experiments***

This type of irradiation experiment is designed to test the whole fuel element (MABB pellet + cladding) in a suitable geometry (not necessarily representative of the future Generation IV SFR technology) so as to realistically reproduce the different temperatures and radial thermal

gradients in the fuel and cladding. These experiments will also study the migration of volatile and gaseous species and any oxide-cladding bond restructuring and/or formation with respect to standard Am-free fuel.

A fast-spectrum experiment will make it possible to reconcile cladding damage with the fuel burn-up. This will broaden the scope of investigation to cover fuel-cladding interactions (corrosion, oxidation, etc.) under thermochemical conditions and thermomechanical loads that are representative of the future SFR technology.

Where possible, these irradiation experiments will be performed in a fast reactor. Otherwise, they will be performed in the MTR with a device capable of screening the thermal component of the flux without overly reducing it. These types of experiments are currently being assessed for reactors such as the ATR (United States), OSIRIS and the HFR in the short term, and for the future JHR in the long term.

#### First semi-integral experiment

Like MARIOS and DIAMINO, it will be necessary to conduct a preliminary experiment to explore the temperature conditions between ~500 and 1 500°C.

The irradiation experiment will be performed under normal operating conditions while aiming for a high burn-up rate compared with the rate targeted for MABB in SFR (at least 50% of the target fission rate which is close to 15 at.% for MABB containing 20% americium).

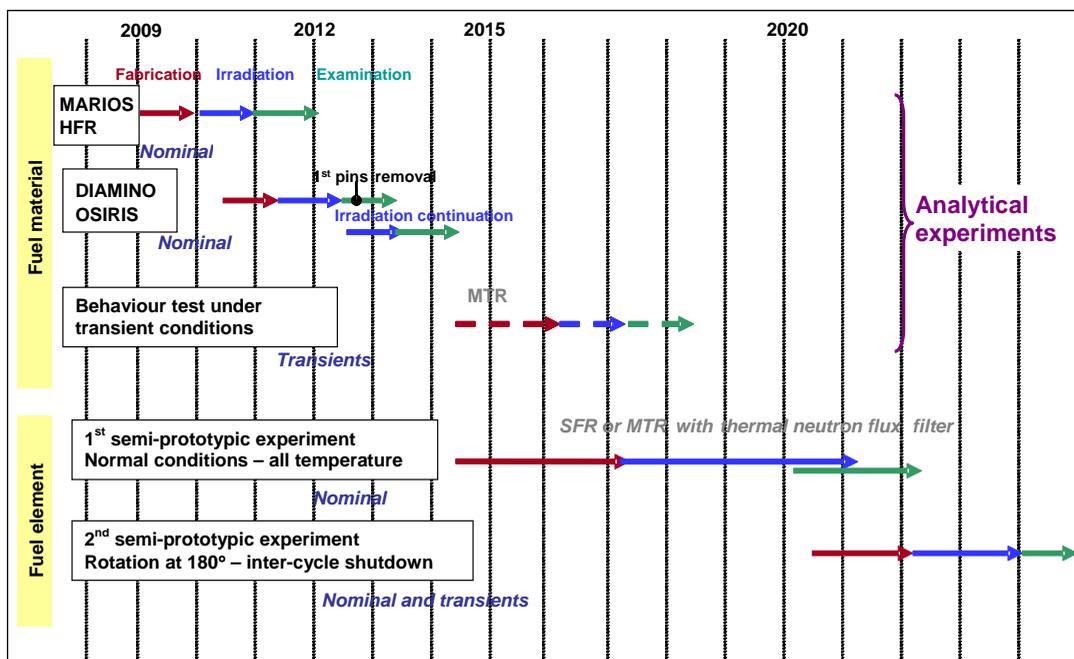
#### Second semi-integral experiment

The second experiment will aim at determining the impact of specific events occurring during the normal irradiation of MABB in SFR, particularly the 180° rotation of sub-assemblies at mid-life and reactor inter-cycle shutdowns.

Ideally, this experiment could follow on from the previous experiment on pins that will have been doubled up and whose irradiation will be prolonged by the study of these transients in the same reactor or elsewhere.

A brief overview of the Phase 2 irradiation experiments is given in Figure 5.

Figure 5: Irradiation experiments scheduled for Phase 2



### Phase 3 – Optimisation

This phase mainly aims to:

- optimise the reference concept and improve performance levels;
- prepare the final specifications for industrial partners and the regulatory documents for the nuclear safety authority;
- validate modelling for off-normal operating conditions;
- guarantee control over the process in the pilot facility and demonstrate that objects can be manufactured in a reproducible manner according to the specifications.

#### ***Integral irradiation experiments***

Compared with the semi-integral irradiations, these integral experiments test the true geometry of the MABB fuel element under representative conditions. Focus is placed on reproducing the axial flux and temperature profiles, as well as the radial thermal gradients in the fuel stack.

#### ***Normal operating conditions***

Integral experiments will first be performed on full pins representing the final concept. These experiments will be conducted in the fast flux of the ASTRID prototype reactor. The pins required for the irradiation programme will be manufactured in the ALFA facility which should be commissioned in 2025.

The first tests will be performed under normal operating conditions up to an actinide fission rate which will be very close to the nominal rate targeted in SFR (around 15 at.%). The pins will be subjected to intermediary examinations which require doubling the number of pins for each irradiation condition under investigation. The MABB sub-assembly 180° rotation at mid-life will also be reproduced for the pins which will be maintained up to a maximum burn-up. The main goal in this case will be to control the potential factors limiting the lifespan of the MABB pins. The most conservative conditions in terms of behaviour (particularly the irradiation temperature) will have to be modelled and applied during these tests.

If the transmutation of all the MA (Am + Np + Cm) is still deemed relevant at this time, several pellets containing Np and Cm may be inserted into the fissile columns of several pins for irradiation in this type of integral experiment. Several full MABB pins may also be loaded into a MABB bundle.

The impact of manufacturing variations in the fuel element characteristics (pellets and cladding) upon behaviour during irradiation should be quantified during this optimisation phase. The goal is to achieve the target operational values by cumulating all the possible margins of variation in the manufactured objects.

#### ***Incident conditions***

The second tests will assess safety and the impact of the different incident and accident transients in the core upon the MABB fuel elements. These tests may be performed on irradiated whole pins or pin sections reconditioned into instrumented mini-pins to be irradiated in an experimental reactor.

#### ***Modelling***

The code used to model the fuel element will be completely validated for normal and off-normal operating conditions at the end of this phase. The nuclear safety authority must be shown that all the phenomena capable of occurring during any of the above conditions have been covered by modelling.

## Phase 4 – Qualification

The objective of this phase is to:

- demonstrate the manufacturing process on an industrial scale from the fuel material through to the sub-assembly;
- qualify the production line to be set up;
- confirm the correct behaviour of the sub-assemblies under incident and accident conditions;
- demonstrate core safety;
- validate modelling up to the scale of the sub-assembly.

As the ASTRID prototype will be available during this phase, the sub-assembly qualification tests could be performed in this reactor. A fabrication facility with a greater capacity than that of the ALFA project will have to be commissioned to manufacture whole sub-assemblies.

An irradiation test on a whole sub-assembly could be performed up to operating levels exceeding the nominal conditions targeted for this technology. This would guarantee its correct operation in the specified operating range.

A specific monitoring programme will have to be set up for this sub-assembly qualification operation.

## Conclusion

With a view to optimising the irradiation experiments, this MABB development programme provides a framework for identifying and sequencing the scientific and technical programmes which support the design studies for the fuel element through to the sub-assembly. The optimisation phase will be much easier and the experiments better defined if as much progress as possible is made in modelling and simulation. The MARIOS and DIAMINO analytical irradiation experiments are currently being prepared and will be performed in the HFR and OSIRIS reactor, respectively, early 2011 and in mid-2012. These experiments required manufacturing MABB samples in the hot lab of the Atalante facility. Two different microstructures were made with the objective of checking their impact on fuel behaviour during irradiation.

This programme will be updated regularly in relation to:

- strategic orientations and decisions resulting in design or size changes;
- progress of technical and scientific programmes;
- availability of experimental means to manufacture objects and perform irradiations;
- availability of materials and manufacturing means.

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## Fuels for the United States DOE Deep Burn programme\*

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### Abstract

Recent irradiation testing in the United States has demonstrated excellent performance for tristructural isotropic (TRISO) coated uranium oxide/uranium carbide fuel kernels designed for use in high-temperature gas-cooled reactors. These same TRISO coatings can also be applied to fuel kernels containing transuranic (TRU) elements from used nuclear fuel. This TRU-TRISO fuel is capable of going to high burn-up to dramatically reduce the total inventory of transuranics in the spent fuel. The United States Department of Energy Deep Burn Project was initiated at the end of 2008 to establish the fundamental understanding and fabrication capability demonstrated by high burn-up TRU-TRISO fuel. We will report on progress towards development of an internal gelation process for the production of the TRU kernels followed by fluidised bed chemical vapour deposition of the TRISO coatings. Additionally, modelling and surrogate kernel fabrication technology demonstration for internally gettered fuels, using SiC nanoparticles, will be presented.

Current work in 2009 and 2010 involves the use of surrogate materials in place of the transuranic elements. The paper will also discuss the status and plans for glove box operations slated to commence at the end of 2010 to allow for the introduction of the transuranics, with the goal of fabricating the first of the TRU-TRISO fuel by the end of 2011.

Work was sponsored by the United States Department of Energy's Office of Nuclear Energy.

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\* The full paper being unavailable at the time of publication, only the abstract is included.



## Progress of the CEA ALFA project: Atalante Laboratory for Actinide-bearing Fuel Manufacturing

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### Abstract

Since 2009, the French CEA has been working at the Marcoule site on a project devoted to design in the Atalante facility a new laboratory known as the Atalante Facility for Actinide-bearing Fuel Manufacturing (ALFA). As part of the CEA waste management programme which aims to assess the feasibility of minor actinide transmutation, the main objective of this project is to design and build a facility dedicated to the fabrication at a significant demo scale of minor actinide (Np, Am and Cm) bearing fuels and blankets.

In the field of MA fuel development programmes, the objective is the optimisation and technological validation phase at pin scale.

Under this framework, the overall need for irradiation qualification tests of MA-bearing fuel has been evaluated and leads to a yearly MA oxide production line capacity between 1 and 5 kg. ALFA will be able to produce various MA bearing fuel and blanket types. The MA concentrations will vary between a few per cent (homogeneous recycling) to a maximum of some tenths of per cent (heterogeneous recycling)

The criteria for the design of ALFA have been defined as follows:

- *Productivity: reducing the fabrication lead time for experimental elements to a few months (currently years).*
- *Flexibility: fast and easy adaptation of the process for various reactor specifications.*
- *Innovation: demonstration of innovative technologies in the architecture and the operation of hot cells, as a demo mock-up of what could be a future facility at a significant scale.*

A preliminary ALFA optimisation plan has been studied, including a global roadmap for an investment plan having an objective of beginning production in 2025 (to be confirmed).

This new facility will be opened to international collaboration.

## Introduction

The Atalante Facility for Actinide-bearing Fuel Manufacturing (ALFA) project calls for the construction of an experimental high-activity fuel fabrication facility to address the R&D requirements for synthesising and producing the elements necessary for future experimental irradiations, in order to define, characterise and qualify fuel for advanced fuel cycles, especially minor actinide loaded fuels.

## Scope and objectives of the project

The objective is to provide a basis for a decision concerning the construction at Marcoule of an experimental high-activity fuel manufacturing facility. It must have sufficient capacity to manufacture a few fuel pins each year with a fissile column a few tens of centimetres long, up to experimental pins for the future JHR and ASTRID reactors (with fuel characteristics constituting the design basis for the production of the ALFA project).

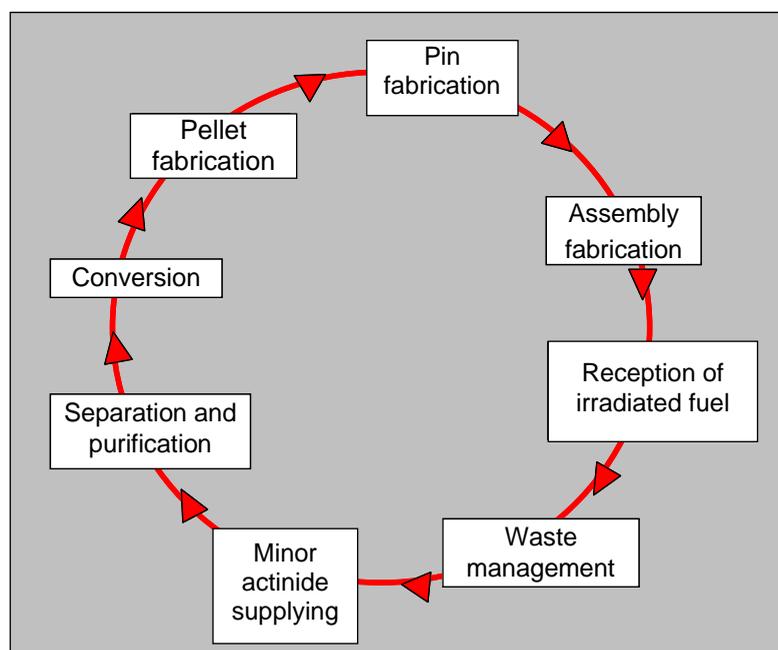
It must be as flexible as possible to meet French and international needs. ALFA will allow the fabrication of a range of fuels for different types of reactors.

With the objective of a subsequent transition to industrial operation, the facility will be required to demonstrate control of the fabrication process with tools validated at significant scale, and allow testing of innovative layout and remote handling concepts to facilitate tool maintenance, operation and upgrading.

As the facility will subsequently close the R&D on future fuel cycles (including experimental fuel treatment), the layout design must take into consideration the available resources in Atalante; some functions can be performed outside the facility, and others can be shared. Figure 1 describes the different functions to be performed.

The project consists therefore of three main units focusing on oxide powder production, pellet fabrication and cladding.

**Figure 1: ALFA perimeter**



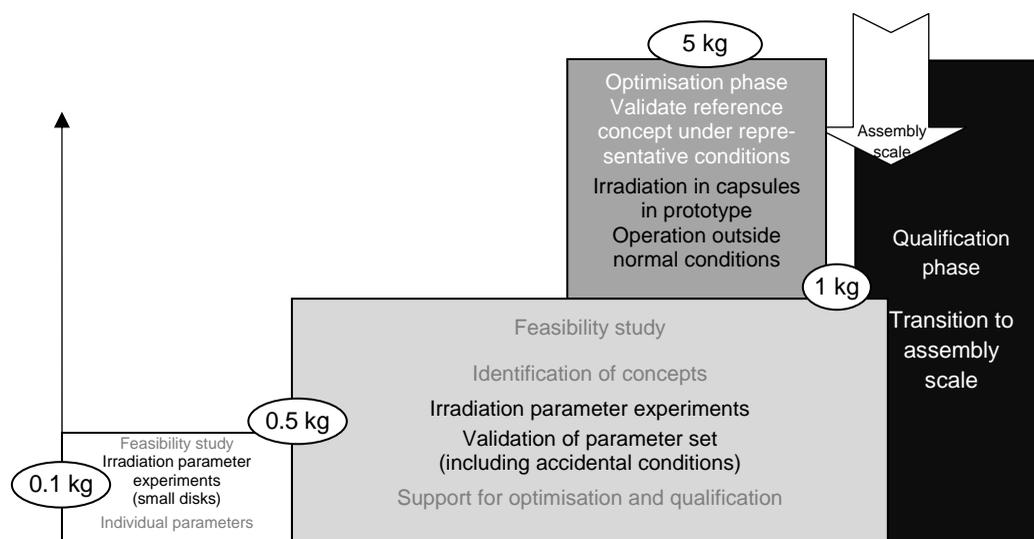
## Main requirements

### Production capacity

A requirements analysis for the ALFA facility was performed to develop minor-actinide-bearing blanket fuel for the ASTRID reactor, chosen as a representative case study to evaluate an order of size for the production capacity (as shown in Figure 2). A development programme would ideally include four development phases: Phase 1: Fuel and core design, Phase 2: Feasibility, Phase 3: Optimisation, Phase 4: Qualification. The mass range of oxide fuel containing MA can be estimated for these phases as follows:

- First part of Phase 2 (white zone): 100 to 500 grams/year.
- Second part of Phase 2 (light grey zone): 500 to 1 000 g/year.
- Phase 3 (dark grey zone): 1 to 5 kg.
- Phase 4 (black zone): qualification of a fuel assembly would require quantities of material incompatible with a working zone in Atalante.

**Figure 2: Development phases**



The first feasibility step (white zone) could be initiated today with the existing resources; this is not the case for the second feasibility step (light grey zone). The ALFA project should therefore focus on the light grey zone with provisions for future development to meet the needs of the dark grey zone. In other words, ALFA should focus on ensuring a responsive fabrication capacity (a few months total fabrication time including characterisation) for pellets, pins and capsules if necessary. In conclusion the facility will have to be optimised to produce objects for irradiation with an annual capacity of 1 kg of powder and fuel pellets, the design including the possibility of a maximum annual capacity of 5 kg. Those values will be consolidated with the irradiation programme under definition.

### Product specifications

The different specifications of fuel for transmutations are still under definition and will be consolidated thanks to the first part of irradiation programme, especially as concerns the knowledge for the appropriate pellet microstructure.

Concentration of MA will be of a few per cent for minor-actinide-bearing fuel up to some tenth per cent for minor-actinide-bearing blanket as shown in Table 1.

Table 2 illustrates the various geometrical characteristics of objects to be produced for different reactors.

**Table 1: Fuel composition**

	Fuel composition
Minor-actinide-bearing fuel <b>Homogeneous mode</b>	U = 77 to 85%; Pu = 14 to 19%; AM = 1 to 5%
Minor-actinide-bearing fuel <b>Heterogeneous mode</b>	U = 70 to 90%; AM = 10 to 30%

**Table 2: Objects' characteristics**

	ASTRID	MONJU	JOYO			JHR
<b>Pins or rods</b>						
Height (mm)	2 741	2 813	1 000	1 500	1 533	1 000 max (w/o instrumentation)
Diameter (mm) (ID/OD)	11.23	5.56/6.5	7.5	6.5	4.8/5.5	<10
<b>Fissile columns</b>						
Fissile column height (mm)	1 200	930	200	200	500	600 mm
Fissile column mass (g)	≈950	220	≈70	≈50	≈90	≈450
Number of pellets	≈120	≈120			≈60	≈100 max
<b>Pellets</b>						
Type	Annular	Cylindrical	Type I Cylindrical	Type II Cylindrical	Fuel Cylindrical	Experimental capsule
Diameter (mm)	10/2 (OD/ID)	5.4	6.54	5.42	4.63	
Height (mm)	~10	8			9	100-200 (to be confirmed)
Mass (g)	≈8 g	≈1.8 g			≈1.6	

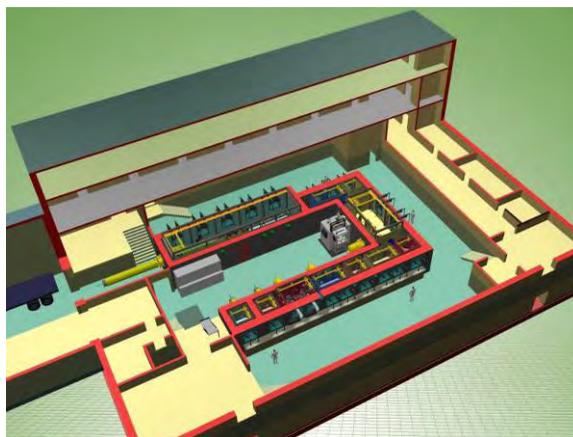
## Studies progress

An evaluation of the available resources in the Atalante facility displayed the possibility to achieve the liquid process (separation purification and conversion processes) in the existing building and the need for a new building (Figure 3) for the dry process (pellets, pins and capsules fabrication). A first design (Figure 4) of the fabrication line has therefore been performed based on well known existing PWR MOX or SFR fabrication process to provide a first estimation in terms of realisation and cost of an investment plan.

This first design already takes into account some innovations such as telerobotics and varied technologies allowing rapid reconfiguration and maintenance of critical equipment, but a consolidation of these options is under evaluation.

Generally speaking the specificities of minor actinides (high activities, neutron emissions, criticality risk and thermal effects) will have an impact on the operating process conditions and on the fabrication equipment in the hot cell environment. Moreover the use of powder metallurgy to produce oxide fuel could be the initial design reference process for the ALFA facility; an evaluation of alternative processes must be done.

That is why the ALFA project is tightly linked to the development of the two main R&D programmes.

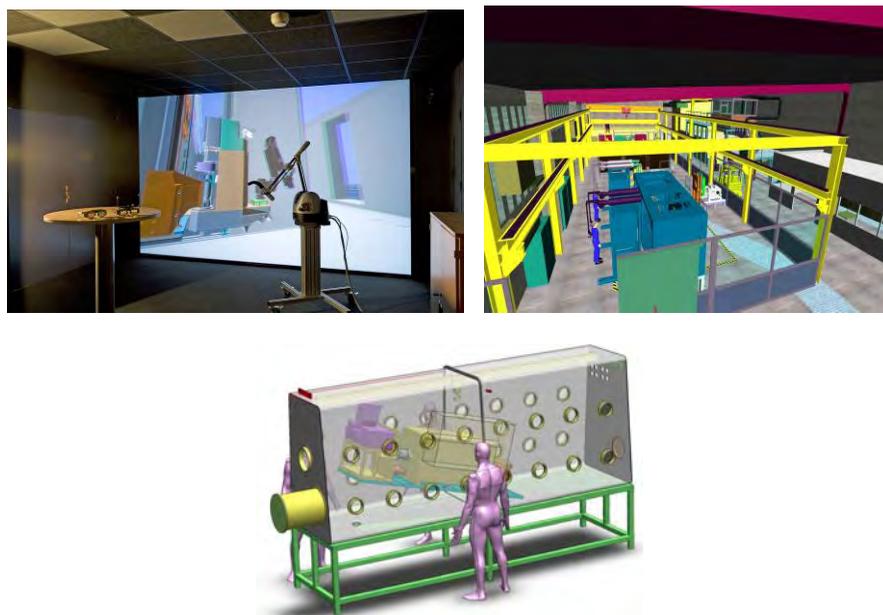
**Figure 3: Artist view of the new building****Figure 4: Fabrication line**

After the selection of adapted routes for conversion and fabrication, thanks to multi-criteria analysis, the first programme aims to achieve the developments allowing the validation of the chosen process at a significant scale. For example we can mention for the short or middle term the direct oxalic co-conversion route, the weak acid resins for the fabrication of microspheres allowing direct pressing and sintering of the pellet or for long-term development, a more innovative process such as SPHEREPAC.

The second programme concerns technological development. The objectives consist in the validation of innovative options for hot cell architectures, for example the last developments in telerobotics and viewing systems open the way to several new approaches allowing a better use of the available space in the hot cell that will be constrained only by the accessibility of the robots [use of robot controlled remotely by a force-feedback master arm with the robot device mounted on a travelling device in the cell; replacement of the current generation of telemanipulators by robotic manipulators on which the master arm is no longer mechanically linked to the slave arm (technology tested at AREVA's La Hague reprocessing plant)]. In the same way many innovation aspects are required for the development of process technologies. The equipments currently used in the fuel cycle for operations similar to those planned for ALFA (powder preparation, pelletising, sintering) are not directly applicable to the fabrication of MA-bearing fuel. The constraints arising from the high activity (radiation, heat, contamination, use of robots or telemanipulators) require a completely new design.

Those development works will focus on operability, maintainability and flexibility and will be performed thanks to the implementation of simulation tools for hot cell facility design such as platforms, mock-up and virtual reality (Figure 5).

**Figure 5: Virtual reality, equipment test platform in inactive area, uranium qualification**



## Conclusion

The ALFA project calls for the construction of an experimental high-activity fuel fabrication facility to address the R&D requirements for synthesising and producing the elements necessary for future experimental irradiations for transmutation. ALFA will be able to produce various MA-bearing fuel and blanket types. The proportion of minor actinides (Am, Np and Cm) will be significant (with a maximum of some tenth %).

As such ALFA will be a significant enhancement of the Atalante facility in order to address the global R&D needs for the advanced fuel cycle.

The roadmap to be confirmed aims to define the preliminary design of the project in 2014 and starting production from 2024.

This new laboratory will be opened to international collaboration as an international user facility.

## Behaviour of zirconium on passivated U-Zr-based fuel alloys

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### Abstract

*X-ray characterisation of annealed fuel pellets revealed an increase in the  $\alpha$ -Zr content on the surface (X-ray active depth) of the fuel that is not representative of the expected equilibrium fuel phases. It has been suggested that  $\alpha$ -Zr is stabilised by the presence of oxygen. Since there is some oxide content on a passivated fuel surface, the oxide ions can migrate into the fuel during vacuum heat treatment while zirconium migrates to the fuel surface. Zirconium can also phase-separate near the fuel surface. We investigated this behaviour with preliminary modelling and with X-ray diffraction. We compared: i) a prior heat treatment of passivated fuel pellets with ii) a new heat treatment of fuel slugs that were subsequently pelletised. The XRD pattern for a new heat-treatment – on fresh surfaces of U-Zr-based fuel pellets that were cut from annealed slugs – did not show increased  $\alpha$ -Zr on the sample surface. Since U-Zr-based EBR-II type fuels are known to give a zirconium surface rind that offers some resistance to fuel-cladding chemical interaction, an understanding of this behaviour is important for optimal fuel performance when fabricating in a hot cell facility with low O<sub>2</sub> levels. This could also prove to be important depending on the fuel fabrication method used (e.g. if oxide coatings are not used in a casting process). Further characterisation work is planned.*

## Introduction

Recent experimental results have provided insight into the interaction between the zirconium in U-Pu-Zr alloys and dissolved oxygen. Other researchers have noted that  $\alpha$ -zirconium is stabilised by the presence of dissolved oxygen [1, 2], and our findings agree. Moreover,  $\alpha$ -zirconium can be stabilised just from vacuum-annealing ternary alloys with an ordinary passivation layer of oxygen on the alloy surface as reported here. While X-ray diffraction patterns for as-cast alloys do not show the presence of  $\alpha$ -Zr, vacuum-annealed samples do exhibit significant levels of  $\alpha$ -Zr even though the equilibrium phase diagram does not predict it for the composition ranges studied. As a consequence, additional samples were prepared using another method in order to understand this behaviour.

For transmutation fuel applications, understanding the behaviour of zirconium on the fuel alloy surface can be important for controlling or enhancing fuel performance. It is believed that a zirconium rind on the alloy surface reduces fuel-cladding chemical interaction (FCCI). The zirconium rind arises partly as a result of casting the fuel alloy slug in a silica-glass mould with a wash coat of  $ZrO_2$  in aqueous suspension. The intent of the  $ZrO_2$  wash coat was to allow an easy and clean separation of the alloy fuel slug from the silica-glass mould. The side benefit for FCCI was thought to be significant enough to try an irradiation experiment with some fuel alloy slugs cast in thin zirconium moulds [3].

In the event that transmutation fuels are prepared in a shielded hot cell, it is possible that the entire cell atmosphere is made up of argon. Under that condition, any passivation layer that forms for a transmutation fuel alloy slug will be different from the passivation layer that ordinarily forms for fresh fuel exposed to air. As a result, the fuel performance with regard to FCCI may actually be different and possibly worse. To help address this concern, we investigated some of the characteristics for  $\alpha$ -Zr formation on vacuum-annealed, air-passivated alloys of U-Pu-Zr.

## Background

Zirconium is added to U-Zr and U-Pu-Zr metallic nuclear fuels to raise the solidus temperature. The different behaviours of zirconium relate to the phenomena influencing its behaviour. One characteristic mentioned above arises from using a wash coat of zirconia in the silica-glass moulds for casting the alloys. A second characteristic arises from fuel constituent redistribution under the thermochemical gradients along the radius of the fuel slug [4, 5]. A third characteristic is possible owing to the stabilisation of  $\alpha$ -Zr for low oxygen contamination levels within the fuel. For the purpose of this discussion, further distinction of these three behaviours may be useful.

Aqueous suspensions of zirconia have been used to coat silica-glass moulds prior to casting the fuel slugs. At the high temperatures for casting (in excess of 1 200°C), some chemical equilibration is possible between zirconia and uranium metal, so that a fraction of the uranium is oxidised to urania while the zirconia is reduced to the metal. (In fact, zirconia is slightly more stable than urania so that some equilibrium exchange is favoured.) As a consequence, a zirconium “rind” can form on the fuel slug, and such a rind has been characteristically observed for U-Zr alloy castings that were prepared for EBR-II. This so-called rind is *not* the focus of this paper.

During irradiation of U-Zr-based fuels in EBR-II, the temperature gradient along the fuel radius (centreline to surface) spans approximately 80°C. This thermal gradient and the variation in thermodynamic stabilities of fuel alloy phases at different temperatures promote the redistribution of fuel constituents [2]. Under irradiation, zirconium gets depleted midway along the radius and gets enriched at the centreline *and* at the fuel slug surface [3]. While the zirconium does slightly concentrate at the peripheral region of the fuel slug, this region is presumably very broad (approximately 1 mm) compared to surface phenomena that would be dominated by a passivation layer (oxide layer) that is present when the fuel is inserted into the reactor. The fuel constituent redistribution in U-Pu-Zr alloys is also *not* the focus of this paper.

Low oxygen impurity levels (e.g. 1 000-2 000 ppm) in U-Zr and U-Pu-Zr alloy systems have been reported to stabilise  $\alpha$ -Zr phase for alloy compositions in which  $\alpha$ -Zr is not indicated as one of the equilibrium phases [6]. Moreover, the presence of oxygen in concentrations greater than 100 ppm was found to enhance the compatibility of the steel cladding and fuel phases [7]. Note that the oxygen stabilisation of  $\alpha$ -Zr can contribute to the rind formation during casting as described above, and it is possible that the presence of oxygen and the concomitant presence of  $\alpha$ -Zr in the rind are the bigger contributors to the reduction of FCCI. Presented here are the preliminary results for oxygen-stabilised  $\alpha$ -Zr on the surface of U-Pu-Zr fuel alloys. While this surface behaviour *may* be a factor in rind formation on the fuel slug (as caused by using a ZrO<sub>2</sub> wash coat on the silica-glass moulds), the  $\alpha$ -Zr formation and stabilisation is being addressed here separately from rind formation during casting.

## Experimental

Six U-Pu-Zr alloy compositions were cast and examined using X-ray diffraction and SEM on solid pellets. These alloy compositions were chosen to be cast for a variety of reasons, including the characterisation of prospective low-fertile metallic fuels with a conversion ratio as low as 0.25, higher TRU content alloy with conversion ratios close to unity but not a breeder, etc. In the course of performing the thermal and phase characterisations of these alloys, it was decided to address the behaviour of oxygen-stabilised  $\alpha$ -Zr with variables that include heat, ambient air and vacuum. Of these six alloy compositions, two compositions – Alloy 1 and Alloy 3 – were investigated under a variety of conditions, for which preliminary results are reported here.

## Casting

Alloys for the AFC2-B fuels were constituted by weighing out the raw materials in the proportions required to meet the desired fuel composition (Table 1). Alloying and homogenisation were accomplished by melting the alloy components together using an arc-melting furnace. Each alloy was melted into a button shape, allowed to cool, flipped and re-melted to give at least three melts total. “Ln” in Table 1 is used to designate a lanthanide alloy composed of 53Nd-25Ce-16Pr-6La (in weight %) which is the approximate weight ratio of the major lanthanide fission products.

**Table 1: Designated and measured compositions (wt.%) for six U-Pu-Zr alloys**

Alloy	1	2	3	4	5	6
U	50	25	35	63.5	61	43.5
Pu	30	55	20	20	24	36.5
Zr	20	20	45	15	15	20
Ln	–	–	–	1.5	–	–
Measured (ICP-MS, $\pm 5\%$ at two sigma)						
U	49.6	23.4	33.9	63.4	59.3	41.8
Pu	28.5	52.7	19.1	19.7	23.1	34.9
Zr	19.8	20.1	43.3	15.0	14.4	20.3
Ln	–	–	–	1.45	–	–

Fuel slugs were made using an arc-melting furnace with the drop-casting technique. This process uses a small copper hearth for electrical conduction with a hole in the middle and a ceramic hearth for thermal insulation. This set-up allows enough heat to remain in the melt until the material becomes completely molten (with some super heat) and flows into the silica-glass mould that is located in the hole in the centre of the copper hearth. A new drop-casting hearth is shown in Figure 1.

**Figure 1: A new copper hearth for drop casting**

### **Sample preparation**

For the purposes of this investigation, the main interest was to characterise the cross-sectional surfaces of the fuel slugs with respect to possible differences in zirconium behaviour. While the in-pile FCCI behaviour concerns the interaction between the fuel slug cylindrical surface and the interior cladding wall, those experiments with cross-sectional surfaces are expected to exhibit the same kinds of behaviour that the cylindrical fuel surfaces exhibit prior to contacting the cladding.

Samples of as-cast alloys were prepared for XRD and SEM by taking cross-sections of the fuel slugs (~5 mm diameter) as pellets. Pellets were cut to approximately 1-2 mm thickness using a low-speed diamond-wheel Buehler saw (water-cooled during the cutting). The SEM samples were polished manually on a 3 micron cloth. XRD samples were smoothed manually on 1 200 grit SiC paper.

During annealing of the alloys, samples were held at 500°C for seven days (168 h) followed by natural cooling in the furnace (one day). Annealed samples were prepared using one of two methods. In the first case, the pellets were cut, incidentally exposed to ambient air, and then heat-treated; these are referred to as “pellet anneal” samples. In the second case, a length of fuel slug was heat-treated before sectioning it into pellets to expose a fresh alloy surface; these are referred to as “slug anneal” samples. For these two sample preparations, the pellet anneal samples could show an unusual surface characteristic owing to the passivation layer (oxide layer) present on the pellet surfaces when they went into the furnace for heat treatment. Specifically, the passivation layer is believed to enable the stabilisation of  $\alpha$ -Zr as described here.

### **X-ray diffraction**

X-ray diffraction patterns were taken for the alloys using a Scintag XDS 2000 theta-theta diffractometer. Each XRD sample consisted of mounting three pellets that were cross-sectioned from the alloy slug (~4.1 mm diameter) co-planar to increase signal strength and reduce sampling time. Samples were mounted in a hermetically sealed chamber with a Be window (X1 Environmental Chamber).

LaB<sub>6</sub> [NIST SRM 660a  $a = 4.1569162(97)$  Å] was added to the surface of the pellets as an internal line standard. The measured peak profiles for the line standard were used to correct for

sample displacement and goniometer error. The data were analysed using Rietveld analysis (Bruker AXS, Topas 3.0, Topas 4.2) applying a full-parameter approach (FP). However, the implementation of analytical pseudo-Voigt profile functions (PV) instead of FP functions produced improved fitting of the diffracted X-ray peak profiles for Si standard and fuel phases if the diffracted intensities indicate high crystallinity (small FWHM).

The Rietveld analysis was done in stages. In Stage 1 the line standard was used to correct for sample- and goniometer off-sets. In Stage 2 metallic fuel phases were identified and quantified through peak profile fitting and in Stage 3, oxide structures were included in the Rietveld profile fitting procedure to confirm their presence or absence. Since the samples were not in ideal powder form, it was possible a preferred texture might result. Rietveld analysis as applied to the diffraction pattern allows the correction for preferred orientation by implementing the algorithm of March Dollase, or applying a spherical harmonics calculation. However, preferred orientation of the fuel phases was lower than expected and was not considered for most of the metallic fuel phases of the recorded pattern.

### Microstructure characterisation

Microstructures were examined using the JEOL JSM7000F Scanning Electron Microscope in the Electron Microscopy Laboratory (EML) at the Idaho National Laboratory (INL). Each sample consisted of a pellet embedded in acrylic, polished as described above, and with a thin layer of sputtered Pd to aid in contamination control and eliminate electrostatic charging.

The JSM7000F microscope is equipped with energy- and wavelength-dispersive X-ray spectrometers and is controlled by Oxford Instruments Inca software. Secondary electron (SE) and backscattered electron (BSE) images, energy-dispersive X-ray (EDS) spectra, and wavelength-dispersive (WDS) maps were collected. All data were collected with a nominal accelerating voltage of 20 kV.

## Results

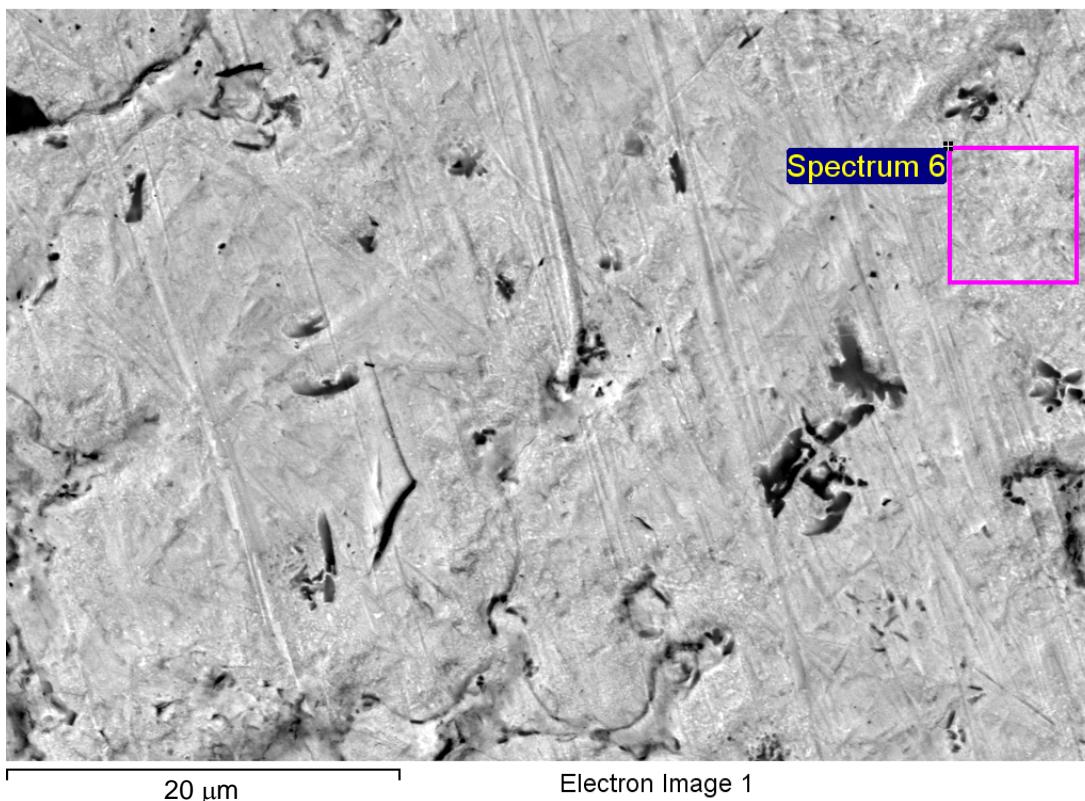
A summary of the XRD results is given in Table 2. Four samples are reported for Alloy 1. The first two entries represent two different castings. There were two castings for Alloy 1; the XRD results give reasonable agreement, considering there is little control over the heating (arc melting) and cooling processes during the casting operation. The pellet anneal samples are pellets that were annealed after they were sectioned from the fuel slug casting (as described above). Slug anneal samples, as noted in the experimental section, arise from annealing slugs and then taking cross-sections of the slugs. Alloy 3 samples included three kinds similar to Alloy 1.

**Table 2: Rietveld refinement results for two alloys (compositions in wt.%)**

Alloy Sample → Condition ↓Phase↓	Alloy 1: 50U-30Pu-20Zr				Alloy 3: 35U-20Pu-45Zr		
	As-cast	As-cast, 2 <sup>nd</sup>	Pellet anneal	Slug anneal	As-cast	Pellet anneal	Slug anneal
α-U	–	–	4.6	31	–	13.8	1.4
γ-U	10	13.2	–	69	–	0.2	0.1
ξ-Pu(U)	39	16.2	82.8	–	–	18.4	1.8
δ-Pu	–	–	–	–	–	4.0	–
δ-UZr <sub>2</sub>	51	70.6	0.3	–	100	6.6	96.8
β-Zr	–	–	–	–	–	–	–
α-Zr	0.2	–	12.3	–	–	57	–

The backscattered electron image for the Alloy 1 as-cast sample is shown in Figure 2. The region marked as Spectrum 6 appeared to be a two-phase mixture with different levels of actinides, which gives agreement with XRD results indicating  $\gamma$ -U and  $\xi$ -Pu(U). X-ray spectra for this region indicated approximately 19 wt.% Zr, 51 wt.% U and 29 wt.% Pu with a small amount of silicon impurity. The dark regions have high zirconium levels corresponding to  $\alpha$ -Zr or  $\delta$ -UZr<sub>2</sub>. The X-ray spectra for the majority of measurements (4 out of 5) of these regions indicate zirconium ranging from approximately 50 to 80 wt.%. It is worth noting this image has high magnification, and that these high zirconium (dark) regions represent a small fraction of the overall cross-sectional surface.

**Figure 2: Backscattered electron image for Alloy 1 as-cast**



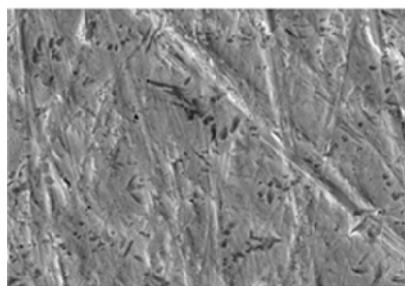
Backscattered electron images (first row) and some of the corresponding X-ray maps (remainder of images) for Alloy 3 are given in Figure 3. The left-hand column of images corresponds to the Alloy 3 as-cast sample section. The right-hand column corresponds to the Alloy 3 slug anneal sample section. SEM of pellet anneal samples are not reported because the one sample looked at thus far had portions that were too friable to handle (as a contamination source). The dark regions clearly have higher zirconium content than the other regions, as the X-ray maps reveal. Except for the dark regions, the X-ray maps for U, Pu and Zr show no distinguishing feature. The same is true for the oxygen X-ray map; however the oxygen X-ray map does not show the high zirconium features, possibly suggesting it is fairly uniform across the surface.

## Discussion

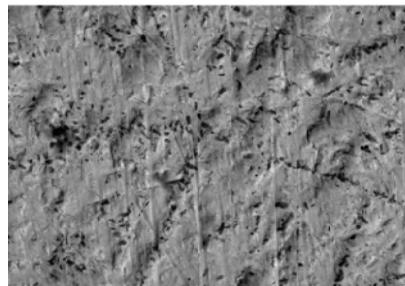
The room temperature phase composition for Alloy 1 is estimated to be a mixture of  $\xi$ -Pu(U) +  $\delta$ -UZr<sub>2</sub> [8], which is in fair agreement with the Alloy 1 as-cast phase composition found by XRD (Table 2). The presence of  $\alpha$ -U and  $\gamma$ -U detected for the Alloy 1 slug anneal perhaps results from

**Figure 3: Backscattered electron and X-ray maps for Alloy 3**

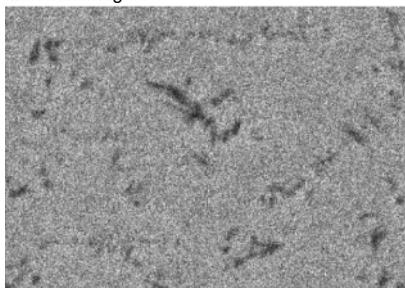
Left column is as-cast sample section, right column is slug anneal sample section



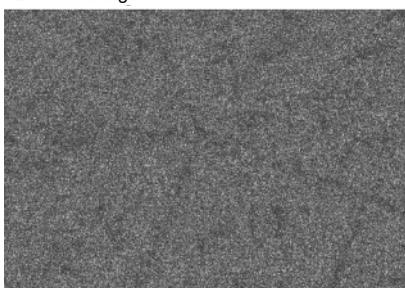
Electron image 1



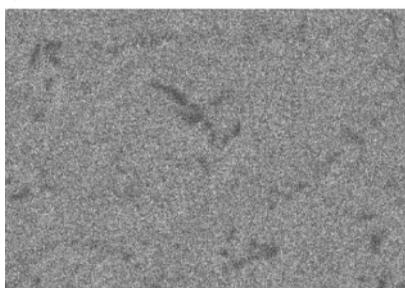
Electron image 1



U Ma1



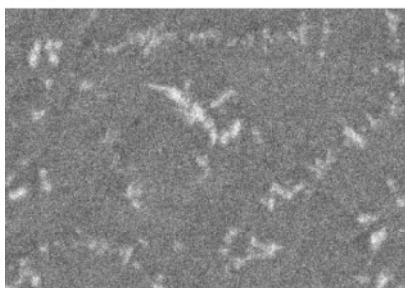
U Ma1



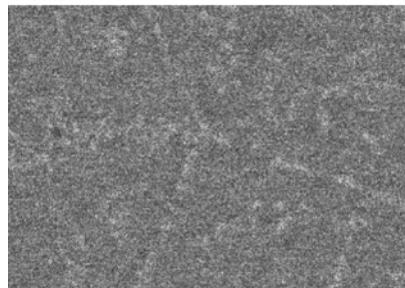
Pu Mb



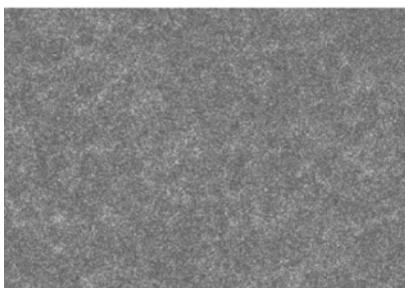
Pu Mb



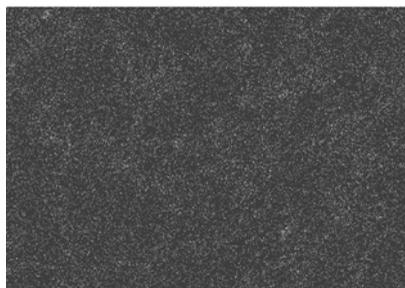
Zr La1



Zr La1



O Ka1



O Ka1

quenching (naturally cooled with a steady argon flow) and a low-temperature transitioning of a portion to the  $\alpha$ -U phase. The room temperature  $\alpha$ -Zr phase is not expected for the Alloy 1 composition.

It is believed from the results of the pellet anneal sample (Table 2) that the  $\alpha$ -Zr phase arises from the heat treatment of pellets that have a passivation layer of oxides. The  $\alpha$ -Zr phase is found for the pellet anneal sample. By contrast, neither the as-cast Alloy 1 sample nor the slug anneal Alloy 1 sample exhibit the  $\alpha$ -Zr phase, even though oxide layers must exist on the samples because they have been exposed to air. Clearly the presence of the  $\alpha$ -Zr in the Alloy 1 pellet anneal samples relates to the surface condition and heat-treatment history. Thus the room-temperature exposure to air is sufficient to produce oxides, and possibly sub-oxides, of zirconium, uranium and plutonium; however, the phase segregation of  $\alpha$ -Zr is apparently inhibited at room temperature.

The room temperature equilibrium phase for Alloy 3 is estimated to be 100%  $\delta$ -UZr<sub>2</sub>. The overall results for Alloy 3 are consistent with those for Alloy 1. The pellet anneal sample exhibits a large fraction of  $\alpha$ -Zr (57 wt.%), which is believed to result from the heat treatment of the passivated pellet surfaces. On the other hand the as-cast sample did not undergo the heat treatment, and the slug anneal sample exposed fresh surface of the pellet to X-ray diffraction. Consequently, the history of the latter two samples does not support formation of the  $\alpha$ -Zr phase, similar to corresponding Alloy 1 samples.

## Conclusion

While these results are preliminary and there is interest to carry out more microstructural characterisation on both types of annealed samples, the XRD results for this investigation demonstrate the influence of sample history on the presence or absence of  $\alpha$ -Zr. The XRD results demonstrate that the  $\alpha$ -Zr is found on samples annealed as pellets ("pellet annealed" samples). On the other hand, the  $\alpha$ -Zr phase is not present in the as-cast sample or in the slug annealed sample, so that it must arise in the pellet annealed sample during the heat treatment. Further, the surveyed literature indicates the  $\alpha$ -Zr is stabilised by the presence of oxygen, so that the passivation layer of oxide on these pellets prior to heat treatment is likely key to the formation of the  $\alpha$ -Zr.

In the event a metallic transmutation fuel alloy is produced entirely in an argon-filled hot cell, then the fuel slugs will not be exposed to ambient air. The passivation layer on the fuel slug is therefore likely to be different from the passivation layer formed on fresh fuel that has been exposed to air. Upon subsequent heating (e.g. in the reactor), the formation of the beneficial  $\alpha$ -Zr on the fuel slug surface as part of the rind will also be different, especially if a ZrO<sub>2</sub> mould wash is not used in the fabrication process. As a consequence, the FCCI for fuel produced entirely in an argon hot cell can be worse than for fuel that has been exposed to air and passivated. Accordingly, this possible difference in fuel performance should be kept in view as progress is made in nuclear metallic fuel alloys for transmutation purposes, and further study is advised.

## Acknowledgements

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## **Session IV**

### **Pyro and Aqueous Processes**

***Chairs: D. Warin, J. Uhlíř***



## ACSEPT – Progress in advanced partitioning and challenges for the future\*

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### Abstract

Actinide recycling by separation and transmutation is considered worldwide and particularly in several European countries as one of the most promising strategies to reduce the inventory of radioactive waste and to optimise the use of natural resources. With its multidisciplinary consortium of 34 partners from 12 European countries plus Australia and Japan, the European Research Project “Actinide Recycling by Separation and Transmutation” (ACSEPT) aims at contributing to the development of this strategy by studying both hydrometallurgical and pyrochemical partitioning routes.

To succeed, ACSEPT is organised into three technical domains:

- i) Considering technically mature aqueous separation processes, ACSEPT works to optimise and select the most promising ones dedicated either to actinide partitioning (for the heterogeneous recycling of actinides in ADS target or specific actinide-bearing blanket fuels in fast reactor) or to grouped actinide separation (for the homogeneous recycling of actinides in fast reactor fuels). In addition, dissolution and conversion studies are under way by taking into account the specific requirements of these different fuels.
- ii) Concerning pyrochemical separation processes, ACSEPT is focused on the enhancement of the two reference cores of process selected within EUROPART with specific attention to exhaustive electrolysis in molten chloride (quantitative recovery of the actinides with the lowest amount of fission products) and to actinide back-extraction from an An-Al alloy. R&D efforts are also brought to key scientific and technical issues compulsory for building a complete separation process (head-end steps, salt treatment for recycling and waste management).
- iii) By integrating all the experimental results within engineering and system studies, both in hydro and pyro domains, ACSEPT will therefore deliver relevant flow sheets and recommendations to prepare for future demonstrations at a pilot level.

After more than two years of work, significant progress has been achieved in process development with the demonstration of SANEX and innovative SANEX flow sheets. Chemical systems were selected for GANEX and are under study. In pyrometallurgy, the work progresses as well. In addition, efforts were made to increase collaborations, mutualise and homogenise procedures and share good practices. Based on these assessments, it is then time to look at the future challenges to be issued.

\* The full paper being unavailable at the time of publication, only the abstract is included.

*A training and education programme is implemented to share knowledge among the partitioning community, and present and future generations of researchers. Specific attention is also given to the funding of post-doctorate fellowships, two having been appointed respectively at the end of 2008 and at the end of 2009. Through this training and education programme, the first ACSEPT International Workshop was organised last March in Lisbon, Portugal. It placed an emphasis on students' contributions (two-thirds of the presentations) and allowed young scientists to meet and exchange with internationally recognised experts.*

## Development of separation process for transuranium elements and some fission products using new extractants and adsorbents

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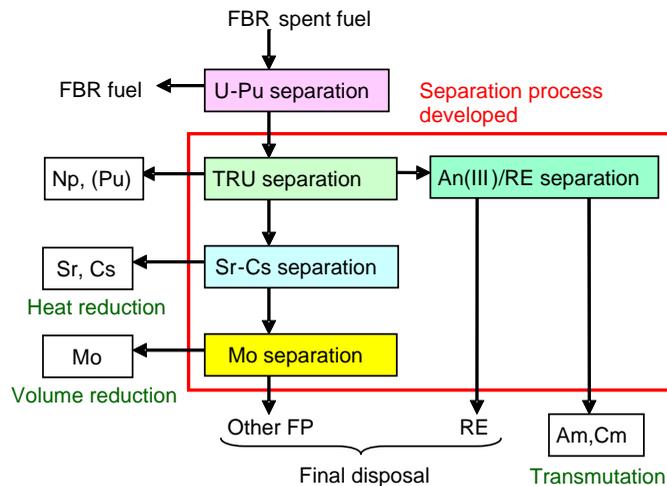
### Abstract

Separation process for transuranium elements (TRU = Am, Cm, Np and Pu) and some fission products (Sr, Cs and Mo) has been developed at Japan Atomic Energy Agency using new innovative extractants and adsorbents to improve the partitioning process from the viewpoints of the economy and the reduction of secondary wastes. Phosphorus-free compounds consisting of carbon, hydrogen, oxygen and nitrogen (CHON principle) were applied to the separation steps for TRU, Cs and Sr by using solvent extraction or extraction chromatography. At the first step, TRU and rare-earth elements (RE) are recovered from high-level liquid waste by solvent extraction with N,N,N',N'-tetradodecyldiglycolamide (TDdDGA). Trivalent actinides Am and Cm, are separated from RE at the next step by extraction chromatography using N,N'-dioctyl-N,N'-diphenylpyridine-2,6-dicarboxamide (Oct-PDA). Heat-generating fission products Cs and Sr are separated from the raffinate of the TDdDGA extraction step by extraction chromatography using calix-crown derivatives for Cs and crown ether derivatives for Sr, sequentially. Finally, Mo is separated by adsorption with an iron oxide adsorbent. This paper presents research and development results concerning the separation process.

## Introduction

A separation process for transuranium elements (TRU = Am, Cm, Np and Pu) and some fission products (Sr, Cs and Mo) has been developed at Japan Atomic Energy Agency using new, innovative extractants and adsorbents to improve the partitioning process from the viewpoints of economy and the reduction of secondary wastes [1]. Phosphorus-free compounds consisting of carbon, hydrogen, oxygen and nitrogen (CHON principle) were applied to the separation steps for TRU, Cs and Sr by using solvent extraction or extraction chromatography. Mo is separated by adsorption with an iron oxide adsorbent. Separation of Cs, Sr and Mo would contribute to the heat and volume reduction of the vitrified waste form of high-level waste. Figure 1 shows the conceptual flow of the separation process developed. The starting solution of the separation process was assumed to be the raffinate of the co-extraction step for U-Pu-Np separation in the NEXT process [2, 3].

**Figure 1: Conceptual flow of the separation process**



We have applied a new extractant, N,N,N',N'-tetradodecyldiglycolamide (TDdDGA, see Figure 2), to the first separation step for TRU (Am, Cm and residual Pu and Np) [4, 5]. TDdDGA is soluble in n-dodecane and has a high extraction capacity with regard to actinides and rare-earth elements (RE). At this first step, TRU and RE are separated from the raffinate mentioned above, which is high-level liquid waste (HLLW), by solvent extraction with TDdDGA. At the next step, trivalent actinide (An(III) = Am and Cm) are separated from RE in nitric acid media by extraction chromatography using N,N'-dioctyl-N,N'-diphenylpyridine-2,6-dicarboxamide (Oct-PDA, see Figure 3) [6]. The Oct-PDA is a very stable compound even when it is in contact with nitric acid at high concentrations. Heat-generating fission products Cs and Sr are separated from the raffinate of the TDdDGA extraction step by extraction chromatography using calix-crown derivatives for Cs and crown ether derivatives for Sr, sequentially, where the nitric acid concentration is kept high, around 3 M. The research and development results of the separation process are presented.

**Figure 2: TDdDGA**

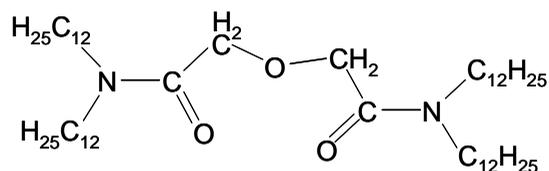
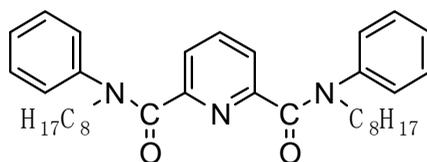


Figure 3: Oct-PDA



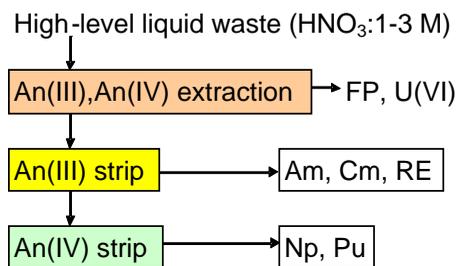
### TRU separation

The new extractant of TDdDGA applied to the separation of TRU and RE from high-level liquid waste gives very high distribution ratio ( $D$ ) at relatively high nitric acid concentration (over 1 M) and low  $D$  at low nitric acid concentration for An(III) and RE [5, 6]. It has a high extraction capacity and can extract Nd at the maximum concentration, one-third of the extractant concentration, without the third phase formation [7].

Among fission products, Zr is extracted by TDdDGA with a very high distribution ratio. Pd itself has a low distribution ratio but is co-extracted with RE [5]. Some complexing agents are required to avoid the extraction of Zr and Pd. Hydroxylethylethylenediaminetriacetic acid (HEDTA) and hydrogen peroxide were selected for this purpose. Hydrogen peroxide functions as a reducing agent for Np(V) and an agent to lower the distribution ratio of Zr.

Tetravalent actinides An(IV), Np(IV) reduced from Np(V) and Pu(IV), are extracted together with An(III). The concept of the first separation step for TRU and RE by TDdDGA is shown in Figure 4. The starting solution contains residual U, Pu and Np, and these three actinides should be controlled. In this flow, An(III), An(IV) and RE are first extracted from HLLW. U(VI) is not extracted and is transferred to the raffinate. Next, An(III) and RE are stripped with diluted nitric acid. Finally, An(IV) is stripped with a complexing agent. Water-soluble DGA compounds can be applied to this stripping step [8].

Figure 4: Conceptual flow of the first separation step for TRU and RE by TDdDGA extraction



A counter-current continuous extraction test using mixer-settler units was performed with simulated HLLW containing 12 fission product elements and  $^{241}\text{Am}$ , where the concentration of RE (Y, La, Nd, Eu) was totally 25.6 mM. More than 99.96% recovery of Am as the product was achieved but some accumulation of Sr in the first mixer-settler was observed [5], which indicates the requirement for further modification of the process conditions.

To avoid Sr accumulation, in each stage of the first mixer-settler the Sr distribution ratio was reduced by increasing the concentration of RE in the organic phase, which was examined using the feed solution at higher concentration of RE and other elements as the next counter-current continuous extraction test (high loading test). The composition of the feed solution in the high loading test is shown in Table 1. The total concentration of RE was 38.4 mM, and the solution also contains  $^{237}\text{Np}$  tracer. Figure 5 displays the operating conditions of two mixer-settler units in the high loading test. As a result of the high loading test, Sr accumulation was not observed as intended. Figure 6 shows the concentration profiles of Am and Np in the first and second

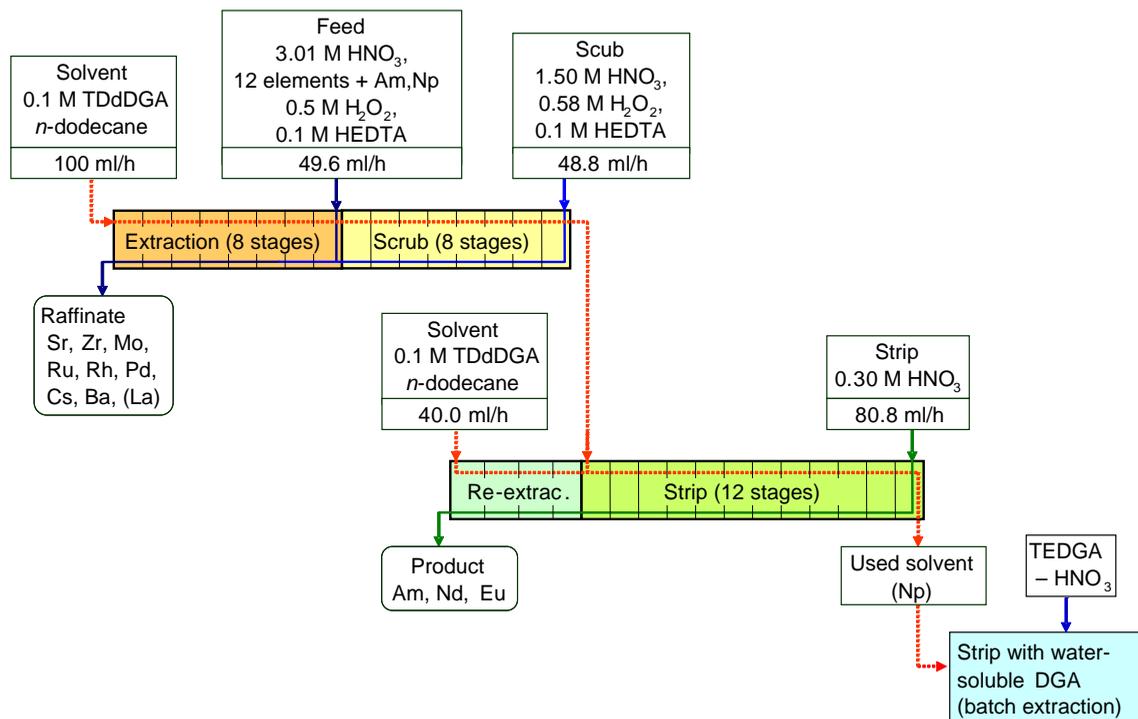
mixer-settler in the high loading test. More than 99.99% recovery of Am from the simulated HLLW was achieved. Np was extracted with a yield of 62%, which would be improved by further addition of hydrogen peroxide. The condition of a stripping step requires further improvement. A simulation method for the extraction process is also being developed, and the calculated concentration profiles of RE agreed well with the experimental results. Optimised process conditions can be given by the calculation with the simulation program.

The solvent from the second mixer-settler was used in the stripping test for Np(IV) and residual elements still contained in the organic phase. The stripping agent used was water-soluble N,N,N',N'-tetraethyldiglycolamide (TEDGA). Efficient stripping of Np(IV) was not obtained with 0.1 M TEDGA-0.2 M HNO<sub>3</sub>, which would be due to the solvent degradation. The stripping test with TEDGA was performed after about one and half months after the mixer-settler test. Quantitative stripping of Np(IV) was demonstrated using the fresh solvent. Residual <sup>241</sup>Am contained in the solvent after stripping with 0.3 M HNO<sub>3</sub> was effectively stripped with 0.1 M TEDGA-0.2 M HNO<sub>3</sub>.

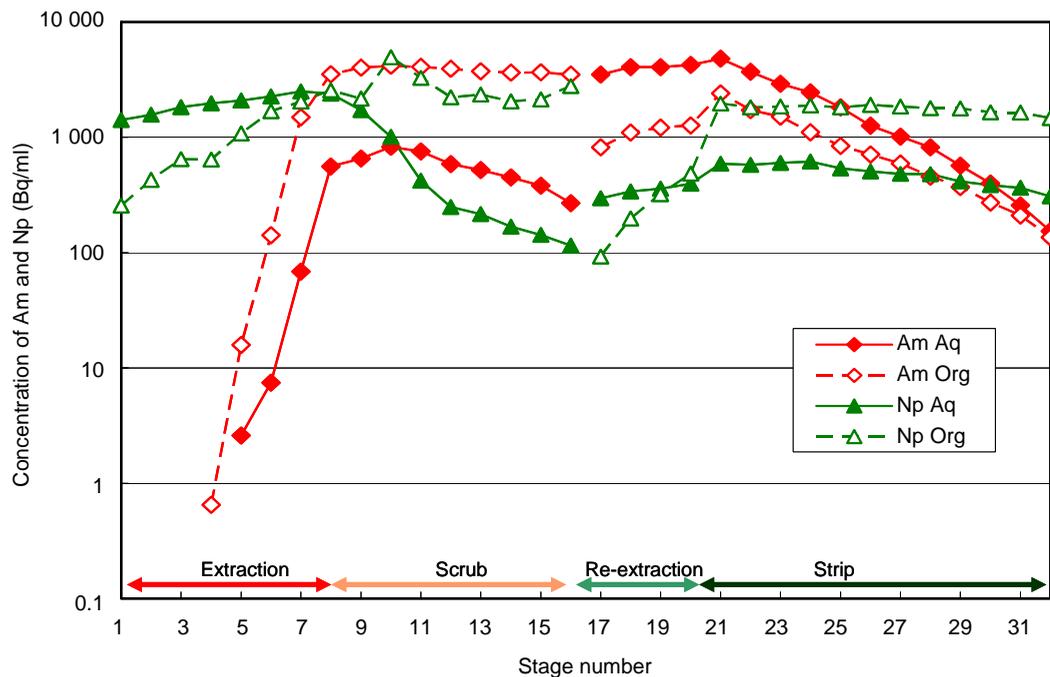
**Table 1: Composition of the feed solution for counter-current continuous extraction test**

Element	Concentration (mM)	Element	Concentration (mM)
Sr	2.48	Pd	11.56
Y	1.39	Cs	14.07
Zr	14.39	Ba	5.87
Mo	9.87	La	11.70
Ru	14.78	Nd	20.75
Rh	4.41	Eu	4.57
HNO <sub>3</sub>	3.0 M	Am, Np	7~8 × 10 <sup>3</sup> Bq/ml

**Figure 5: Operating conditions of two mixer-settler units in the high loading test**



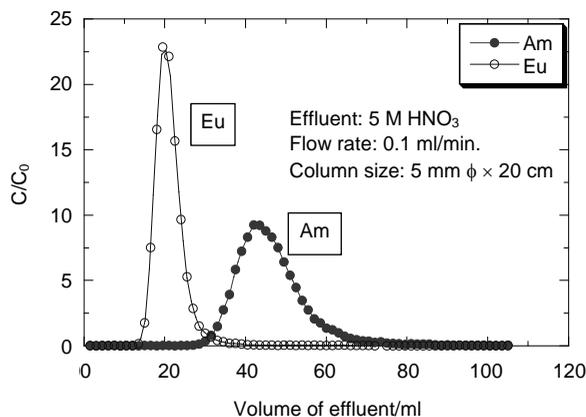
**Figure 6: Concentration profiles of Am and Np in the first and second mixer-settler in the high loading test**



### An(III)/RE separation

After investigation of some pyridine dicarboxamide (PDA) compounds by modifying its alkyl groups attached to amidic nitrogen atoms,  $N,N'$ -dioctyl- $N,N'$ -diphenylpyridine-2,6-dicarboxamide, Oct-PDA (Figure 3), was chosen as the most suitable extractant. The most important property of the PDA extractants was the chemical stability, and they can be used at high concentration of  $\text{HNO}_3$ . The separation factor between trivalent actinide [An(III)] and RE was not so high and therefore Oct-PDA was applied to extraction chromatography for separation of An(III) and RE. Oct-PDA was successfully impregnated into Amberlite XAD-4 resin. The separation of Am(III) from Eu(III) in 5 M  $\text{HNO}_3$  was sufficiently achieved by using an Oct-PDA/XAD-4 column (5 mm  $\phi$   $\times$  20 cm), as shown in Figure 7 [6].

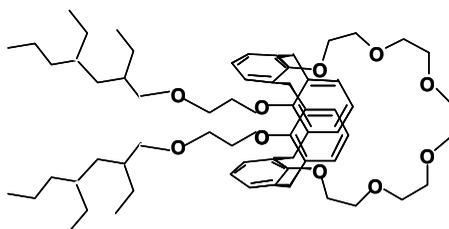
**Figure 7: Separation of Am and Eu by extraction chromatography using Oct-PDA/XAD-4 [6]**



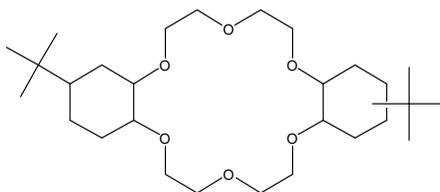
## Sr-Cs separation

Some extractants to separate Cs and Sr from nitric acid solution were developed, but most of them cannot be used in solutions of aliphatic hydrocarbon, such as n-dodecane. Extraction chromatography was, therefore, adopted in the present separation process, wherein those extractants can be used without diluents and Cs and Sr are separated from solutions of relatively high nitric acid concentration. Calix-crown R14 (1,3-[(2,4-diethyl-heptylethoxy)oxy]-2,4-crown-6-calix[4]arene, see Figure 8) and DtBuCH18C6 (di-t-butylcyclohexano-18-crown-6, see Figure 9) were selected for Cs and Sr separation, respectively. Porous silica particles coated with an inert copolymer of formylstyrene and divinylbenzene [9] were used to support the extractants. Both adsorption of Cs with Calix-crown R14 adsorbent and adsorption of Sr with DtBuCH18C6 adsorbent require modifiers. n-Hexadecanol was selected as the best modifier for both Cs and Sr adsorption. The molar ratio of the modifier to the extractant is also one of the parameters to be optimised.

**Figure 8: Calix-crown R14**



**Figure 9: DtBuCH18C6**



Some column adsorption experiments were performed using simulated solutions containing radioactive isotopes where nitric acid concentration was 2 to 4 M [10]. Figure 10 shows one of the results of those experiments. Cs was separated from other elements as an almost isolated fraction. Sr was not adsorbed and was contained in earlier effluent with other elements. Repeated adsorption and elution with simulated solution showed that Calix-crown R14 adsorbent is very stable against extractant elution and displays almost no change in adsorption capacity after 60-cycle repetition.

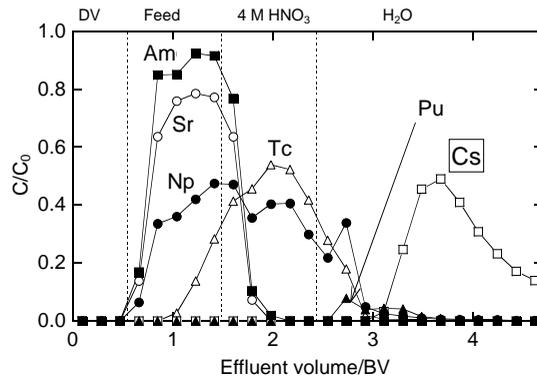
Figure 11 shows Sr separation with DtBuCH18C6 adsorbent [10]. Sr was finally eluted with water along with Tc. Ba also contaminates the Sr fraction. The adsorbent for Sr was less stable compared with the adsorbent for Cs because of the elution of the extractant.

From these experiments, it was concluded that Cs should be separated first and Sr is separated from the Sr fraction in the Cs separation step. An experiment with a genuine HLLW was also performed in a hot cell at the Nuclear Fuel Cycle Safety Engineering Research Facility (NUCEF), JAEA Tokai (Figure 12) and showed almost no change in element behaviour in the Cs and Sr adsorption process.

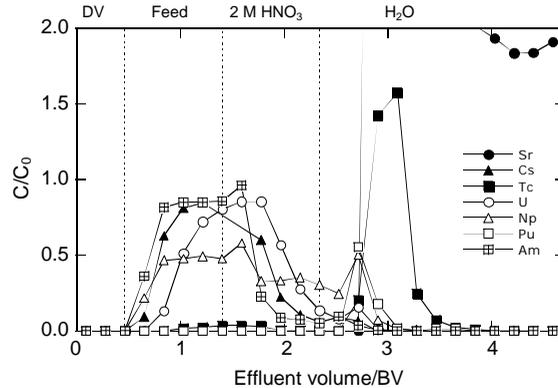
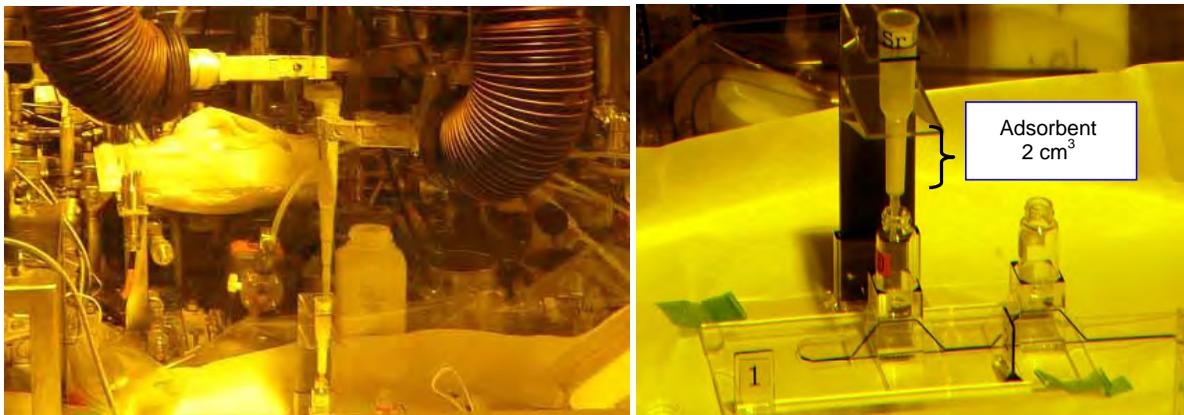
As a final confirmation, a sequential adsorption test using the raffinate of the TRU extraction step with simulated HLLW containing the HEDTA complexing agent was performed with the order of Cs adsorption, Sr adsorption and Mo adsorption (described below). The effluent of Sr fraction from the Cs adsorption column was used as the feed for the Sr adsorption column.

**Figure 10: Separation of Cs through a packed column with Cs adsorbent [10]**

Column: 10 mm  $\phi$   $\times$  240 mm, Calix-crown R14 + *n*-hexadecanol (modifier/extractant = 4.17),  
Flow rate: 1 cm<sup>3</sup>/min, Feed: 4 M HNO<sub>3</sub> solution

**Figure 11: Separation of Sr through a packed column with Cs adsorbent [10]**

Column: 10 mm  $\phi$   $\times$  280 mm, DtBu-CH18C6 + *n*-hexadecanol (modifier/extractant = 1.25),  
Flow rate: 1 cm<sup>3</sup>/min, Feed: 2 M HNO<sub>3</sub> solution

**Figure 12: Small-scale adsorption tests for Cs and Sr separation with a genuine HLLW**

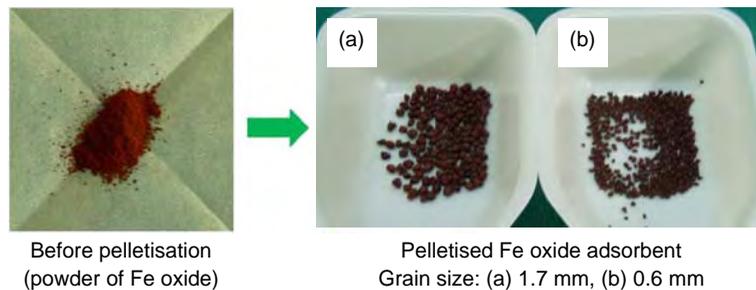
Effluent containing Mo from the Sr adsorption column was used for Mo adsorption. The HEDTA showed no effects on the adsorption of the three elements and almost the same behaviour of elements was observed as the behaviour in the adsorption tests carried out separately. The performance of the separation process series of TRU extraction, Cs adsorption, Sr adsorption and Mo adsorption was verified with the simulated HLLW.

## Mo separation

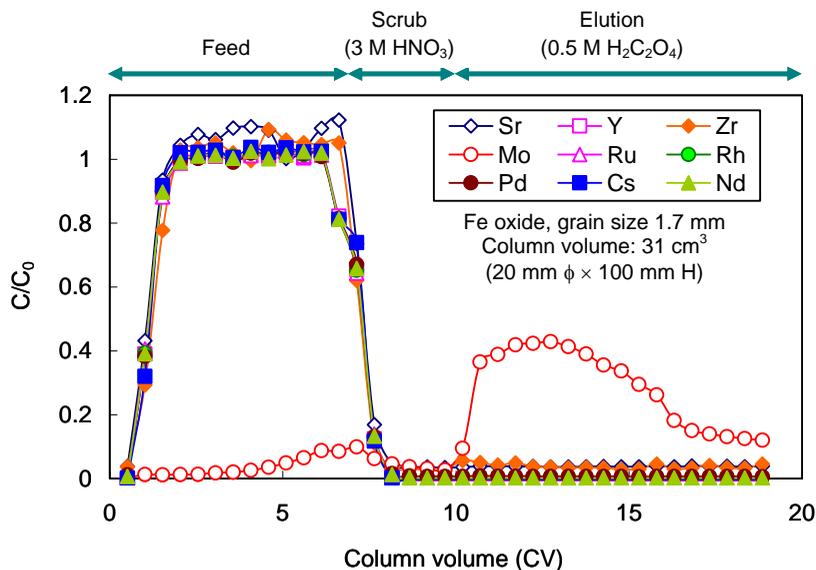
Various metal oxide adsorbents such as Fe-Pd oxide, manganese oxide, alumina oxide, cobalt oxide and amorphous zirconium oxide were examined for Mo separation. Hematite-type iron oxide (Fe adsorbent) was found to have high Mo adsorption ability even in 3 M nitric acid solution and low solubility in nitric acid. TRU, uranium and major fission products were not adsorbed on the adsorbent. Adsorbed Mo can be eluted with an oxalic acid solution.

The adsorbent can be pelletised as shown in Figure 13, which allows the column adsorption. Figure 14 shows the result of a column adsorption experiment. Most of the Mo was separated from the other fission products. The adsorption test with the effluent from the Sr adsorption (mentioned above) showed no difference.

**Figure 13: Pelletisation of the Fe adsorbent**



**Figure 14: Separation of Mo through a packed column with Fe adsorbent using simulated FP solution [11]**

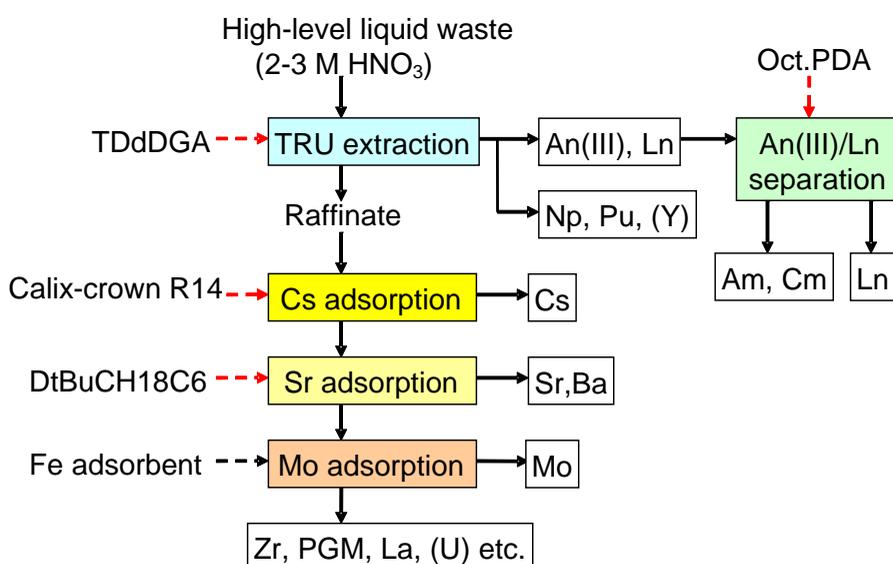


## Conclusion

A separation process for TRU and some fission products was developed using new extractants and adsorbents, which consists of TRU and RE separation by TDdDGA extraction, An(III)/RE mutual separation by extraction chromatography with Oct-PDA, Cs and Sr separation by extraction chromatography with Calix-crown R14 and DtBuCH18C6 respectively, and Mo separation by adsorption with hematite-type iron oxide adsorbent. The flow of the process is shown in Figure 15. All the separation steps were tested using simulated solutions with and without some radioactive isotopes and the Cs and Sr separation steps were verified with a genuine HLLW.

Extraction process for TRU with TDdDGA should be tested with a genuine HLLW as the next step. Further research and development would be required on extraction chromatography from an engineering point of view.

**Figure 15: Separation process flow sheet**



## Acknowledgements

The present study includes results of the study “Development of Separation Technology of Transuranium Elements and Fission Products by Using New Extractants and Adsorbents” entrusted to the Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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## Recent progress in advanced actinide recycling processes\*

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### Abstract

Concerns about environmental preservation have increased the demand for more efficient management, as well as sound and sustainable development, of nuclear energy. Appropriate management of radioactive waste arising at the back end of the fuel cycle is considered to be a crucial issue of long-term environmental concern in relation to the nuclear fuel cycle. Of particular concern – due to long-term radiotoxicity and heat load issues, as well as proliferation risk – is the approximately 0.4 wt.% of spent fuel composed of minor actinide (MA) isotopes. As of the year 2006, it is estimated that about 110 tonnes of MA are being contained in spent fuel storage worldwide, and an additional 40 tonnes are contained in high-level waste products from reprocessing for the future. P&T will be necessary to decrease these amounts.

Within this framework, this paper presents recent progress obtained at CEA/Marcoule on the development of innovative MA partitioning hydrometallurgical processes in support of their recycling, 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 recent tests under high active conditions of 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 also concern 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).

The promising initial results of the EXAm process (extraction of only Am, at a front head step of the partitioning process) are also presented. Since recycling of only the americium could be more easily implemented than the recycling of both americium and curium, EXAm is dedicated to Am recovery at an early stage of the partitioning process, using one hydrometallurgical cycle. The principle of the EXAm process is based on the extraction of americium together with some light lanthanides having close values of distribution coefficients in high nitric acidity, and curium and other lanthanides remaining in the aqueous phase. The TEDGA amide molecule is used in order to increase the selectivities Am/Cm and Am/heavy Ln, because of the complexation of curium and heavy lanthanides by this amide; the overall efficiency of the process is largely improved, with a corresponding decrease in the number of necessary separation stages. A test of this innovative process was carried out at the CEA Atalante facility in 2009, with very satisfactory results, and the concentrations of the radioelements measured during the test are in good

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\* The full paper being unavailable at the time of publication, only the abstract is included.

*agreement with the values calculated with the PAREX code. A complete hot test, from a genuine concentrated spent fuel dissolution solution, is planned in 2011-2012, in order to confirm these promising results.*

*In the coming years, subsequent 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 potential industrial implementation.*

## One-cycle SANEX process development studies and lab-scale demonstrations

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### Abstract

*The direct selective extraction of trivalent actinides from a simulated PUREX raffinate was studied using a mixture of CyMe<sub>4</sub>BTBP and TODGA (one-cycle SANEX). The solvent showed a high selectivity for trivalent actinides with a high lanthanide separation factor. However, the co-extraction of some fission product elements (Cu, Ni, Zr, Mo, Pd, Ag and Cd) from the simulated PUREX raffinate was observed, with distribution ratios of up to 30 (Cu). The extraction of Zr and Mo could be suppressed using oxalic acid but the use of the well-known Pd complexant HEDTA was unsuccessful. During screening experiments with different amino acids and derivatives, the sulphur-bearing amino acid L-cysteine showed good complexation of Pd and prevented its extraction into the organic phase without influencing the extraction of the trivalent actinides Am(III) and Cm(III). The development of a process-like batch extraction series and a continuous counter-current test using a single centrifuge contactor showed very promising results in view of further optimising the process. A strategy for a single-cycle process is proposed within this paper.*

## Introduction

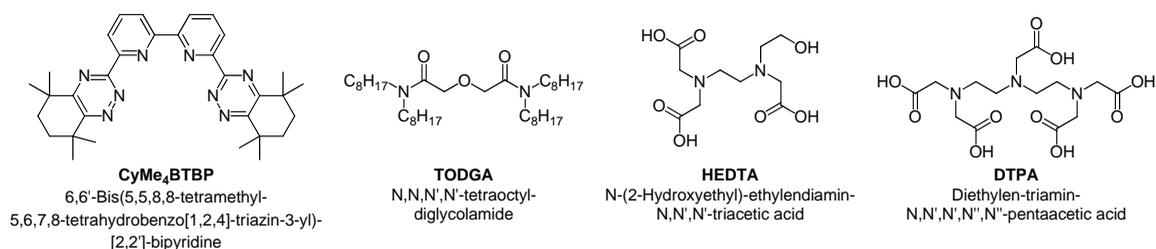
Over the past few decades, two major spent nuclear fuel management options have been favoured by most countries using nuclear power: the direct disposal of fuel elements (open fuel cycle), currently favoured in Germany, Sweden and Finland, and reprocessing (closed fuel cycle), favoured in France and other countries (e.g. United Kingdom, Japan, Russia). Using the Plutonium Uranium Refining by Extraction (PUREX) process, uranium and the fissile plutonium are recovered and recycled as mixed-oxide (MOX) fuel in nuclear power reactors, producing a high active raffinate (HAR), which still contains the long-term radiotoxic minor actinides (Am, Cm) in addition to the fission products. The separation and consecutive transmutation of the minor actinides into short-lived or stable elements may significantly contribute to the reduction of the long-term radiotoxicity of nuclear waste, and thereby to the overall acceptance of nuclear energy production [1].

The development of new and innovative processes for spent nuclear fuel solutions is a very intensively studied topic in nuclear research all over the world [2-4]. As the liquid aqueous waste solution from reprocessing contains approximately 40 different elements in widespread concentrations, the selective separation of trivalent actinides from this multi-element solution is one of the most challenging problems. The separation of the trivalent actinides from the lanthanides is a particularly difficult step, as the two groups of f-elements have very similar physical and chemical properties. This separation is necessary for transmutation because of the large neutron capture cross-sections of many of the lanthanides, which would hamper an efficient transmutation of the actinides.

In Europe, the two-step Diamide Extraction + Selective Actinide Extraction (DIAMEX+SANEX) partitioning process is one of the most promising strategies to be converted from laboratory scale to industrial scale [5-6]. The first step in this process (DIAMEX) uses a diamide extractant to co-extract lanthanides and minor actinides from the highly acidic PUREX raffinate [7-8]. In the subsequent step (SANEX), the trivalent actinides are separated from the lanthanides, e.g. by the highly selective  $\text{CyMe}_4\text{BTBP}$  extractant [9-10]. A drawback of such a strategy is the need for two separate processes using two different extractants. Within the current European project Actinide Recycling by Separation and Transmutation (ACSEPT) [11], a new process design is envisaged: the so-called “innovative SANEX” concept. In this strategy, the trivalent actinides and lanthanides are co-extracted employing a DIAMEX-type process. Then, the loaded solvent is subjected to several stripping steps, involving the selective stripping of the trivalent actinides with hydrophilic complexing agents (e.g. DTPA) followed by the stripping of the trivalent lanthanides [12].

A more challenging route studied within this paper is the direct actinide (III) separation from the PUREX raffinate using a mixture of  $\text{CyMe}_4\text{BTBP}$  and TODGA (structures shown in Figure 1) as extractants. This option is referred to as the one-cycle SANEX process. A single process directly using the PUREX raffinate would reduce the number of cycles, thus saving a co-extraction process (e.g. the DIAMEX process), making the complete advanced reprocessing process easier and more economical.

**Figure 1: Structures of  $\text{CyMe}_4\text{BTBP}$ , TODGA, HEDTA and DTPA**



The  $\text{CyMe}_4\text{BTBP}$  molecule is a highly selective extractant for the separation of actinides from lanthanides [9]. However, the use of a phase transfer reagent is necessary owing to the slow kinetics of the  $\text{CyMe}_4\text{BTBP}$  molecule which can be significantly improved by the use of the

malonamide DMDOHEMA. A successful group separation has been demonstrated in a hot SANEX process using 0.015 mol/L CyMe<sub>4</sub>BTBP + 0.25 mol/L DMDOHEMA in 1-octanol [5]. Modolo *et al.* later proposed substituting 0.25 mol/L DMDOHEMA with 0.005 mol/L TODGA [13]. This system shows comparably good extraction properties and kinetic behaviour compared to DMDOHEMA, and the performance of the system was demonstrated in a spiked test. Within a 24-stage counter-current process, an actinide product fraction containing >99.9% actinides with less than 0.1% lanthanides was obtained. Despite these very promising results, the question arose as to whether it would be possible to directly and selectively extract the actinides using CyMe<sub>4</sub>BTBP from a PUREX raffinate solution, leaving the lanthanides and other fission products in the aqueous phase.

In the present paper, we report the results of batch experiments concerning the possibility of directly and selectively extracting the actinides using a mixture of CyMe<sub>4</sub>BTBP and TODGA. Screening tests for a masking agent for palladium and process development studies including a continuous counter-current test with a single centrifuge contactor using this masking agent were conducted. A strategy for a counter-current process is proposed.

## Results and discussion

### *Extraction from simulated HAR solution*

For the optimisation studies, a synthetic PUREX raffinate solution was used for extraction experiments simulating a raffinate as described in [14]. The relatively high concentrations of Na and Fe in the raffinate are due to purification steps of the U/Pu product produced by the PUREX process and solvent clean-up steps.

On the basis of the reported SANEX process test [5], an extractant mixture consisting of 0.015 mol/L CyMe<sub>4</sub>BTBP and 0.005 mol/L TODGA diluted in TPH/1-octanol = 40/60 (v/v) was used. Table 1 shows the composition of the synthetic PUREX raffinate solution and the corresponding distribution ratios of the elements for the extraction without adding any complexants (Table 1, columns 3 and 7).

These preliminary results show that the direct extraction of Am and Cm from a synthetic PUREX raffinate solution is possible with good extraction of the actinides and a high separation factor of Am/Eu of 68. The distribution ratios of the lighter lanthanides (La, Ce, Pr, Nd, Sm) are even lower, meaning that the separation factor is higher. This is advantageous since the bulk of the lanthanides belongs to the light lanthanides (Table 1).

The trend of increasing distribution ratios with increasing atomic number of the lanthanides for extraction by CyMe<sub>4</sub>BTBP had been observed previously [9]. Also, the distribution ratio reaches a maximum for Dy, and then decreases again.

However, some non-lanthanide fission products are co-extracted with the actinides, namely Zr, Ag, Cd, Mo, Ni, Cu and Pd. Zr and Mo are important because their concentration in the PUREX raffinate solution is fairly high (1 071 and 678 mg/L, respectively). Even a relatively low distribution ratio in this case leads to a considerable loading of the organic phase, thereby reducing the free extractant concentration available for the actinide (III) extraction. Pd (168 mg/L) must also be considered, because of its high concentration in the nuclear waste solution, and it often causes the formation of precipitates, which has to be avoided in a continuous counter-current extraction process. Ag (12 mg/L), Cd (15 mg/L), Ni (40 mg/L) and Cu (19 mg/L) are present at lower concentrations.

The co-extraction of Zr, Mo and Pd is a problem that has often been overcome by the use of the well-known complexing agents oxalic acid and HEDTA (Figure 1) in partitioning processes such as the DIAMEX process [15-16]. Recently we found that the use of HEDTA has no influence on the extraction of Mo and Pd, but only on Zr [17], whereas oxalic acid is a suitable masking agent for Zr and Mo, as shown in Table 1 (columns 4 and 8).

**Table 1: The extraction of actinides and fission products from a simulated PUREX raffinate with CyMe<sub>4</sub>BTBP/TODGA**

Organic phase: 0.015 mol/L CyMe<sub>4</sub>BTBP, 0.005 mol/L TODGA in  
TPH/ 1-octanol = 40/60 (not pre-equilibrated with nitric acid)

Aqueous phase: 3.2 mol/L HNO<sub>3</sub>, HAR elements, complexants given in the  
table + tracers (<sup>241</sup>Am, <sup>244</sup>Cm, <sup>152</sup>Eu), mixing time: 15 min.; T = 22°C ± 1°

Element	Concentration [mg/L or as shown]	Without complexant	C <sub>2</sub> H <sub>2</sub> O <sub>4</sub>	Element	Concentration [mg/L]	Without complexant	C <sub>2</sub> H <sub>2</sub> O <sub>4</sub>
		Distribution ratio				Distribution ratio	
<sup>241</sup> Am	Trace amounts	10.8	9.1	Ag	12	0.88	2.48
<sup>244</sup> Cm	Trace amounts	4.3	3.6	Cd	15	12.3	6.92
Y	90	0.04	0.85	Cr	93	0.02	0.03
La	239	<0.01	0.02	Sn	11	0.12	0.31
Ce	567	<0.01	0.03	Sb	4.6	0.12	0.07
Pr	223	0.01	0.04	Rb	63	0.08	0.10
Nd	718	0.02	0.06	Ru	356	0.09	0.07
Sm	149	0.07	0.14	Rh	73	<0.01	<0.01
Eu	34	0.16	0.24	Te	165	0.03	<0.01
<sup>152</sup> Eu	Trace amounts	0.06	0.22	Sr	177	<0.01	<0.01
Gd	51	0.08	0.17	Ba	259	<0.01	<0.01
Ni	40	30.0	18.4	Cs	542	<0.01	<0.01
Cu	19	4.88	15.7	Al	2	n.d.	n.d.
Zr	1 071	0.50	0.01	Fe	1 900	n.d.	n.d.
Mo	678	2.57	0.21	Se	10	n.d.	n.d.
Pd	168	6.19	8.81	Na	1 600	n.d.	n.d.
HNO <sub>3</sub>	3.2 mol/L						

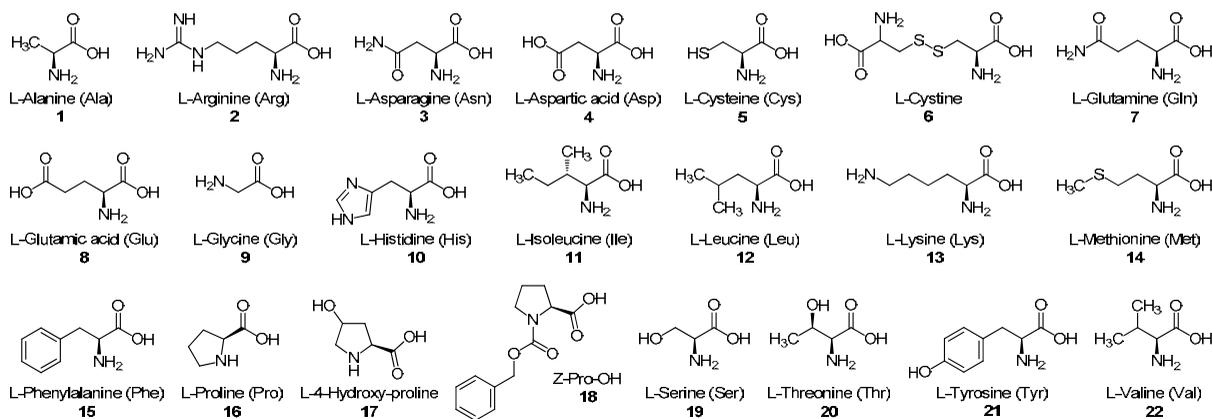
n.d. – not determined.

As Zr and Mo are elements which are present in relatively high concentrations in the synthetic PUREX raffinate solution, the overall loading of the organic phase was significantly reduced. This explains the higher distribution ratios of most of the other elements compared to the experiment without the addition of any complexants. The extraction behaviour of Ag, Cd, Ni, Cu and Pd was not greatly affected by oxalic acid, and the results furthermore show that Y was extracted much better due to the lower overall loading of the organic phase.

### **Screening of different amino acids for palladium complexation**

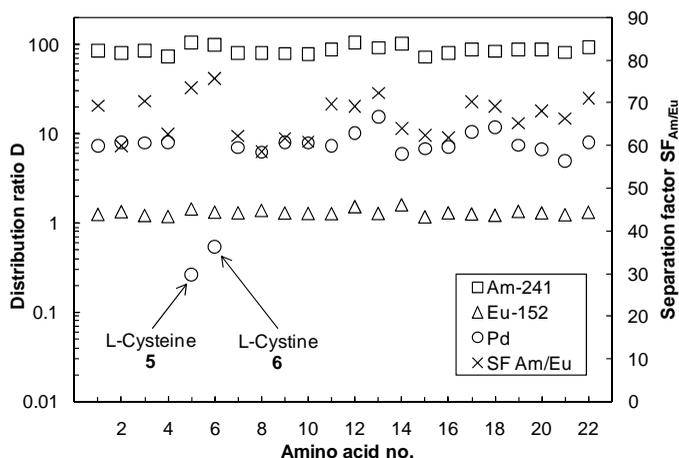
A number of amino acids and derivatives were therefore tested for the suppression of palladium extraction and the influence of the amino acid addition on the extraction of actinides and lanthanides. <sup>241</sup>Am and <sup>152</sup>Eu were used as representatives for the respective groups. An overview of the tested complexants is shown in Figure 2. Amino acids were chosen because of their relatively complex co-ordination chemistry due to the presence of different donor atoms (O, N, S) in diverse structural constitutions as this allows different chelate ring sizes, and because of their good solubility in aqueous solutions. The results of the test with 0.1 mol/L amino acid are shown in Figure 3.

Just two of the 22 tested amino acids showed a significant influence on the distribution ratio of Pd: L-cysteine 5 and L-cystine 6 (structures are shown in Figure 2). The Pd distribution ratios were reduced considerably without influencing the extraction of Am(III) or Eu(III). L-cysteine was selected for further study because of the structural similarity of the two molecules; it was shown that the complexation of Pd by L-cysteine is independent of the nitric acid concentration in partitioning-relevant concentrations (0.1-3.0 mol/L) and that an effective complexation can be achieved by a three-fold excess of the L-cysteine concentration over the Pd concentration [17].

**Figure 2: Overview of the tested amino acids and derivatives****Figure 3: Amino acid screening during the extraction of Pd, Am (III) and Eu (III) by the CyMe<sub>4</sub>BTBP/TODGA solvent**

Organic phase: 0.015 mol/L CyMe<sub>4</sub>BTBP + 0.005 mol/L TODGA in TPH/1-octanol = 40/60 (not pre-equilibrated with nitric acid)

Aqueous phase: 3 mol/L HNO<sub>3</sub>, 150 mg/L Pd (0.0015 mol/L), 0.1 mol/L of different amino acids + tracers (<sup>241</sup>Am, <sup>152</sup>Eu, <sup>244</sup>Cm), mixing time: 15 min.; T = 22°C ± 1°C



### Process-like extraction series experiments with simulated HAR solution

In the following batch experiments, a process-like extraction series was used, comprising an extraction step, two scrubbing steps and one stripping step. After completing each step aliquots of each phase were taken for analysis and the loaded organic phase from this step was subjected to the next step where it was contacted with freshly prepared aqueous phase. The composition of the aqueous phases (Feed, Scrub 1, Scrub 2, Strip) is shown in Table 2.

**Table 2: Composition of the aqueous phases used in the first experiment**

	Step 1: Ex	Step 2: Scrub I	Step 3: Scrub II	Step 4: Strip
c(HNO <sub>3</sub> )/mol/L	3.2	1.0	1.0	0.01
c(oxalic acid)/mol/L	0.3	0.2		
c(L-cysteine)/mol/L	0.05	0.05		

During this first extraction experiment, L-cysteine caused a voluminous precipitation. Analysis of the supernatant solution showed that the precipitate mainly consisted of Pd, Ag, Cd and Ba. Therefore the composition of the aqueous phases in the extraction series was altered, as shown in Table 3. L-cysteine was not added to the feed or the first scrub solution, but was introduced during the second scrubbing step. With this set-up, the production of a separate Pd raffinate would be possible. In a counter-current process, an extra outlet for this Pd raffinate would be inserted, concurrently removing the L-cysteine from the aqueous phase to avoid precipitation.

**Table 3: Composition of the aqueous phases used in the second experiment**

	Step 1: Ex	Step 2: Scrub I	Step 3: Scrub II	Step 4: Strip
c(HNO <sub>3</sub> ) / mol/L	3.2	1.0	1.0	0.01
c(oxalic acid) / mol/L	0.3	0.2		
c(L-cysteine) / mol/L			0.01	

During this experiment, no precipitate formed and no third-phase formation was observed. Figure 4 (left) shows that Am and Cm were well extracted and that they stayed in the organic phase during the scrubbing steps. In the last step (Strip), they were back-extracted into the aqueous phase with distribution ratios below 0.01 for Am and Cm. Eu was poorly extracted, and therefore a high Am/Eu separation factor of 50 was obtained in the extraction step. Even in the subsequent scrubbing steps, the Am/Eu separation factor reached values above 20. As shown in Table 1, Eu belongs to the best-extracted lanthanides, and thereby even higher Am/Ln separation factors were obtained for the other lanthanides. The good back-extraction behaviour of the CyMe<sub>4</sub>BTBP system is advantageous for the development of a continuous process with recycling of the organic phase which could be reused after a possible solvent treatment.

**Figure 4: Distribution ratios and separation factor of <sup>241</sup>Am, <sup>244</sup>Cm and <sup>152</sup>Eu (left) and organic phase concentration of some stable elements determined by ICP-MS (right) in a simulated four-step process using HAR simulate solution**

Organic phase: 0.015 mol/L CyMe<sub>4</sub>BTBP + 0.005 mol/L TODGA in TPH/1-octanol = 40/60  
(not pre-equilibrated with nitric acid; in the other steps, the loaded phase from the preceding step was used)

Aqueous phase: as shown in Table 3, mixing time: 15 min. each step

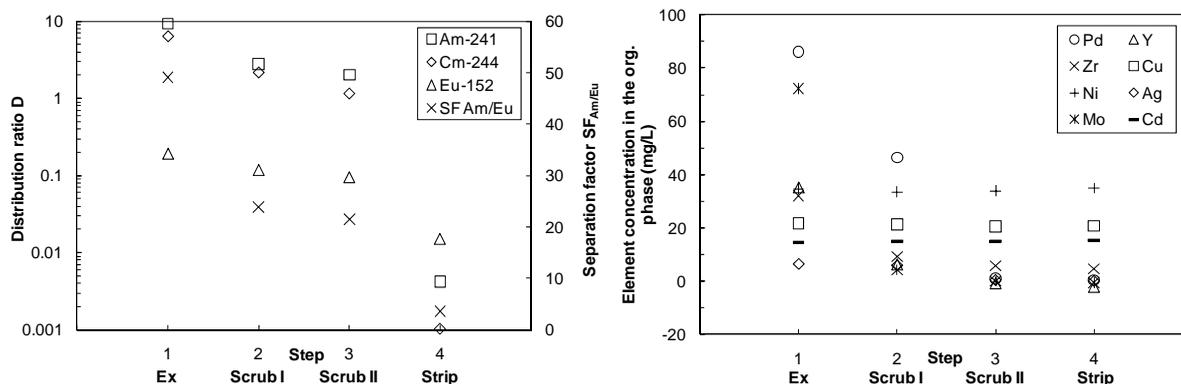


Figure 4 (right) shows the results of ICP-MS analysis for some selected inactive elements, which were co-extracted to some extent. In Figure 4 (right), the concentration of elements in the organic phase is shown instead of the distribution ratio because the D-values of Cu, Cd and Ni could not be precisely determined due to their low concentration in the aqueous phase. The results show that Cu, Ni and Cd are nearly completely extracted, and that they stay in the organic phase during the whole experiment. They could possibly be scrubbed in an alkaline solvent treatment, but this topic was not investigated during this work. The extraction of Zr and

Mo was prevented by the use of oxalic acid, and Y was scrubbed during the first two steps. Furthermore, the results show that Pd and Ag were effectively scrubbed from the organic phase in the third step, which involved the addition of L-cysteine.

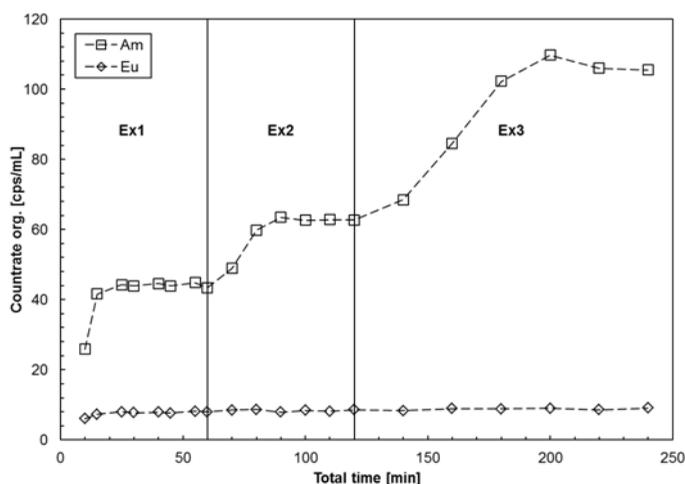
### Continuous counter-current tests using a single centrifuge contactor

Continuous counter-current tests using a single centrifuge contactor were conducted for three steps, selective actinide (III) extraction, Pd-scrubbing and stripping of the actinides. For the extraction test a mixture of HAR feed solution with the first scrub solution was used, resulting in an overall  $\text{HNO}_3$  concentration of 2 mol/L and an oxalic acid concentration of 0.25 mol/L. Freshly prepared solvent was used and the test was run at three different flow rates:

- org./aq. = 20/40 mL/h (Ex1);
- org./aq. = 10/20 mL/h (Ex2);
- org./aq. = 5/10 mL/h (Ex3).

Figure 5 shows the Am and Eu concentrations in the organic phase against the experiment time. Approximately 3-4 exchanges of the mixing chamber volume (ca. 5.5 mL) were needed to reach a steady state.

Figure 5: Am(III) and Eu(III) concentration in the organic phase during the extraction test



The results shown in Figure 5 and Table 4 demonstrate that the stage efficiency increases with decreasing flow rates. For further scrubbing and stripping tests a flow rate of 10 mL/h for both the aqueous and organic phase was chosen. Like in the batch experiments, co-extraction of Cu, Ni, Cd, Ag and Pd was observed to a larger extent and minor co-extraction of Y. The lanthanides follow the same trend as observed previously and Mo and Zr were sufficiently masked by oxalic acid.

Table 4: Results of the extraction test

	Flow rate org. [mL/h]	Flow rate aq. [mL/h]	Distribution ratio		SF
			Am	Eu	Am/Eu
Ex1	20	40	0.85	0.18	4.7
Ex2	10	20	1.38	0.17	8.1
Ex3	5	10	5.72	0.21	27
Equilibrium value by shaking the content of the mixing chamber after Ex3			12.3	0.23	53

The scrubbing test using L-cysteine showed a decrease of the Pd distribution ratio of 2.46 (Ex2) to 0.34 during the scrubbing test. The equilibrium value of 0.03 was not reached due to the slow kinetics of the system. Cu, Ni and Cd quantitatively stayed in the organic phase and were not scrubbed. The slightly co-extracted elements Mo, Ru, Y and the lanthanides were further scrubbed in the aqueous phase.

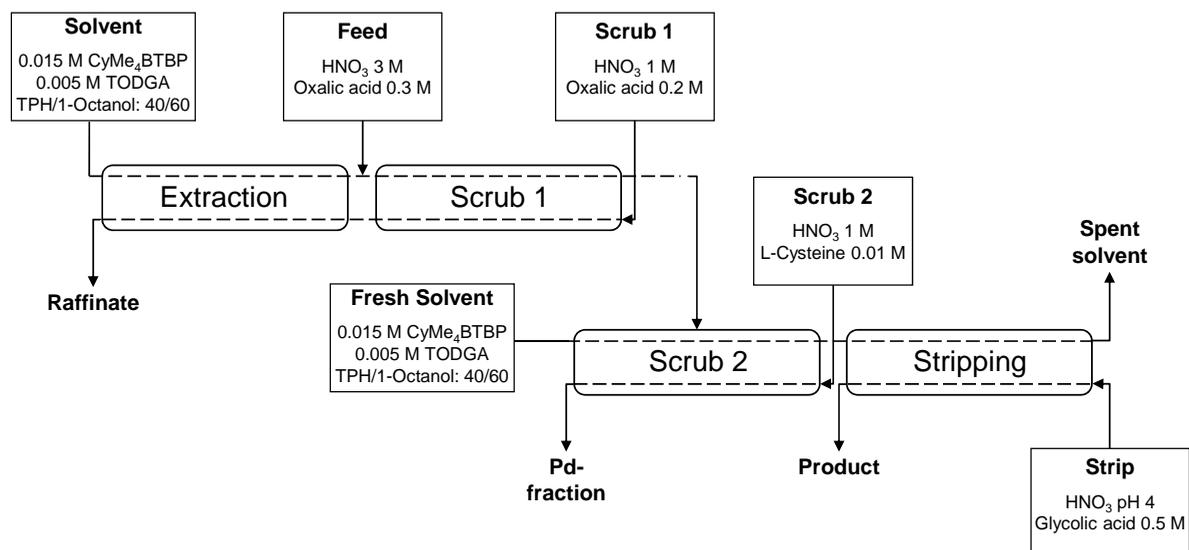
For the stripping test, different aqueous phases were used. First a solution of glycolic acid (0.5 mol/L), set to pH 4 with  $\text{NH}_3$  (St1) was used, then the aqueous phase was changed to a nitric acid solution of pH 3 (St2) and after that to pH 2 (St3).

**Table 5: Results of the stripping test**

	pH	Complexant	Distribution ratio	
			Am	Eu
St1	4	0.5 M glycolic acid	0.11	0.08
St2	3	–	0.74	0.15
St3	2	–	0.67	0.13
Equilibrium value by shaking the content of the mixing chamber after St3			0.01	0.01

The best stripping results were obtained with 0.5 mol/L glycolic acid at pH 4. Thermodynamic stripping at pH 3 or even pH 2 was possible but equilibrium values were not reached owing to the slow kinetics of the system. Cu, Ni and Cd stayed in the organic phase during the stripping test, thus being effectively separated from americium. The product fraction, besides americium, contained smaller concentrations of Pd, Ru, Y, Mo and some lanthanides. These contaminations should be avoided using a higher number of stages for the extraction, scrubbing and stripping parts. A possible flow sheet of a spiked one-cycle SANEX process is shown in Figure 6.

**Figure 6: Flow sheet proposal of a spiked one-cycle SANEX process**



## Conclusions

In this work, it was shown that the direct and selective extraction of trivalent actinides from a synthetic PUREX raffinate solution as one-cycle SANEX seems to be possible. Distribution ratios for Am(III) and Cm(III) were high and their separation from the trivalent lanthanides and most of the other fission and corrosion products was also very high. Nonetheless, there was co-extraction

of some of the non-lanthanide fission product elements in the process candidate, namely Zr, Ag, Cd, Mo, Ni, Cu and Pd. The extraction of the most abundant fission products, Zr and Mo, was suppressed satisfactorily by the use of 0.3 mol/L oxalic acid. The extraction of the other elements was not influenced by the use of oxalic acid or 0.05 mol/L HEDTA.

Due to the high content of Pd in the PUREX raffinate solution, measures were studied to suppress the extraction of Pd. Various amino acids and derivatives were tested and L-cysteine showed good Pd complexation behaviour, whereas the extraction of trivalent actinides and lanthanides was not affected maintaining the high selectivity of the system.

In preliminary process development studies we could demonstrate that the direct selective extraction of trivalent actinides from a synthetic PUREX-type solution is possible and L-cysteine could be used as a selective Pd stripping agent.

Continuous counter-current experiments using a single centrifuge contactor gave very promising results and a flow sheet for a spiked test was proposed.

During the studies it was noted that the co-extraction of Ni, Cd and Cu must be taken into account for further process development studies. The elements are not stripped back to the aqueous phase, which could possibly lead to their accumulation in the solvent. More investigation is required to examine possible stripping conditions for the regeneration and reuse of the spent solvent.

### Acknowledgements

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## Development of a novel GANEX process

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### Abstract

The waste from nuclear power plants has to be isolated from man and his environment for about 100 000 years to be considered safe. It has been suggested that if the long-lived actinides could be separated from the spent fuel and transmuted, the isolation time could be shortened to about 1 000 years. This, however, requires selective separation of parts of the waste. Partitioning and transmutation research in Europe has for the major part taken place within several European Union Framework Programmes. Within the projects NEWPART, PARTNEW and EUROPART a process scheme for the partitioning of nuclear waste from the PUREX process was developed. The scheme includes the DIAMEX, SANEX and SESAME-processes among which both the DIAMEX and SANEX processes have been successfully tested on genuine spent fuel. However, in the latest EU project ACSEPT, which started in 2008, another approach towards partitioning is being investigated. This is the so-called Group Actinide Extraction (GANEX) process. In the GANEX process all the actinides in the dissolved spent fuel are extracted as a group and hence separated from the lanthanides as well as the rest of the fission and corrosion/activation products. A novel GANEX process has been developed at Chalmers University of Technology in Sweden. This new process utilises the properties of already well-known extractants by combining BTBP and TBP into one solvent. The system is able to extract U, Np, Pu and Am from strong nitric acid and simultaneously separate these elements from the lanthanides. This is done with sufficiently high distribution ratios and separation factors without the need for any redox control.

The concept of a GANEX process would bring large benefits industrially by implementing only a very few extraction steps compared to the extraction scheme PUREX-DIAMEX-SANEX-SESAME previously investigated. The novel GANEX extraction process developed at Chalmers University of Technology in Sweden meets many of the basic demands necessary for a viable partitioning process, indicating that GANEX could be a possible future process alternative.

## Introduction

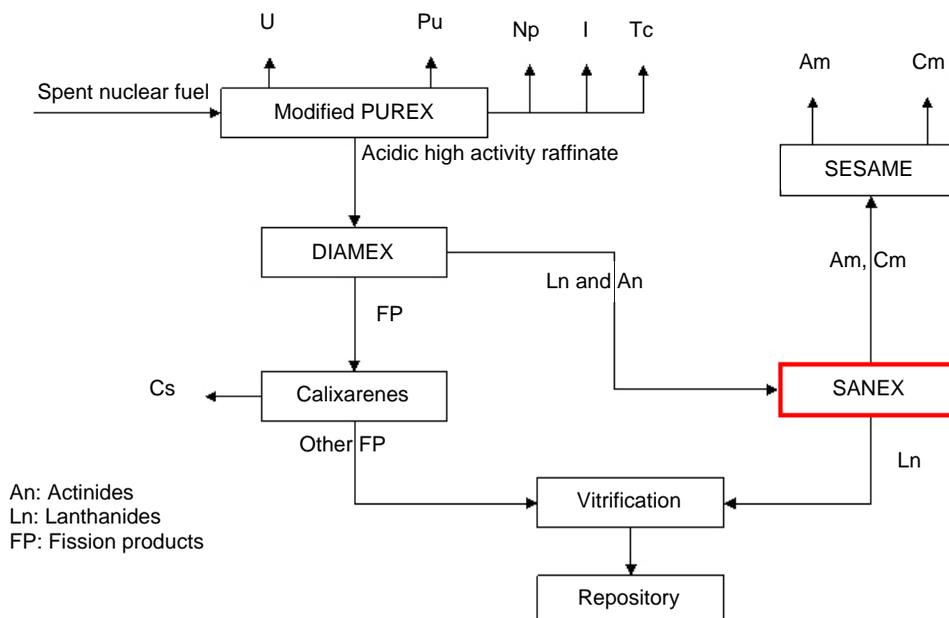
Spent nuclear fuel is one of the main problems associated with the use of nuclear power. Many countries have selected the so-called “once-through” cycle where the spent nuclear fuel is treated as a waste and disposed of in final repositories, often underground. These repositories are planned to keep the waste isolated from contact with humans or other biotopes for more than 100 000 years. An option to this handling is the reprocessing aimed at recovering the uranium and plutonium from the waste in order to make new fuel, so-called mixed-oxide (MOX) fuel. However, even in this case the remaining nuclides such as neptunium, americium and curium require a very long storage time. In recent years the idea of partitioning and transmutation has become increasingly popular. The main idea is to take the remaining solution after the reprocessing and then separate out the long-lived elements for further irradiation and fission into shorter-lived fission products. By applying this strategy the storage time of the nuclear waste would have been reduced to less than 1 000 years and the energy utilisation increased [1].

Over the years several processes and flow sheets for separation for transmutation have been designed, e.g. TRUEX, TALSPEAK, TRPO, DIDPA, UNEX, DIAMEX and SANEX. Of these, DIAMEX and SANEX have been developed in Europe and are discussed in detail below. However, this chapter deals with the development of a fairly new concept, the so-called Group Actinide Extraction (GANEX) process. The main idea behind it is, as the name suggests, to simultaneously remove all the actinides from dissolved spent fuel after the bulk part of the uranium has been removed. This renders a separation of the actinides from the lanthanides as well as the rest of the fission and corrosion/activation products in one operation. The actinides can then be selectively stripped for transmutation purposes [2].

## Extraction process development in Europe

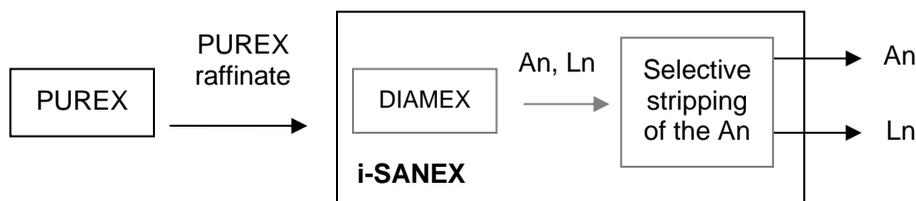
During the different EU projects mentioned above, the strategy for the partitioning work in Europe has changed its focus as progress within the area has been made. During the EUROPART project a full extraction scheme for the reprocessing process was developed [3]. The scheme that can be seen in Figure 1 was developed over many years and several projects.

**Figure 1: Extraction scheme for the partitioning of spent nuclear fuel [5]**



In the i-SANEX process actinides and lanthanides are firstly co-extracted into an organic phase with a di-amide (e.g. TODGA) just like in the DIAMEX process. Secondly however, there is a selective back extraction of the actinides with a hydrophilic complexing agent, separating them from the lanthanides. For a schematic explanation of the process see Figure 2.

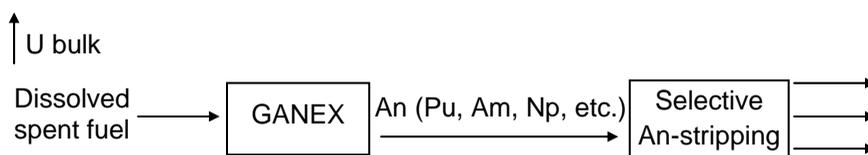
**Figure 2: Schematic picture of the i-SANEX process**



As can be seen in the figure above, the i-SANEX process is quite similar to the American reversed TALSPEAK process. This process is however quite complicated and therefore not well understood and difficult to control [4].

The GANEX process (see Figure 3) is today under development in at least three different ways among the members of ACSEPT. The rest of this article will however only deal with the Swedish model developed at Chalmers University of Technology in Gothenburg.

**Figure 3: Schematic picture of the GANEX process**



### Development of a novel GANEX extraction system

The Chalmers idea behind the new GANEX system was to combine two well-known extractants that display complementing properties, i.e. extraction of trivalent actinides and hence separation from the trivalent lanthanides as well as extraction of tetra- penta- and hexavalent actinides. With these properties of the extraction system there would be no need for redox control of e.g. Pu. This ternary system consists of cyclohexanone as the diluent and the two extractants BTBP and TBP. TBP already has a well-documented ability to extract uranium and plutonium directly from dissolved spent fuel [5], while BTBP molecules on the other hand are known to be able to extract trivalent and pentavalent actinides and hence separate them from the trivalent lanthanides [6, 7].

When starting development of the extraction process, three basic demands need to be met:

- 1) The organic system has to be able to withstand the strong acidic conditions present (an acidity of 4 M HNO<sub>3</sub> has been set as a standard for GANEX experiments within the ACSEPT project) when extracting directly from dissolved spent fuel.
- 2) The system has to be able to extract all of the actinides.
- 3) The system has to be able to separate all the actinides from the lanthanides.

If the system is found to meet these demands and is therefore worth investigating further, then several other important questions have to be considered:

- 4) Can the system perform the extractions and separations after having received a large dose of radiation?

- 5) Does the system extract any fission- or corrosion/activation products?
- 6) How does the system perform under the heavy metal loading conditions that will exist when extracting directly from dissolved spent fuel?

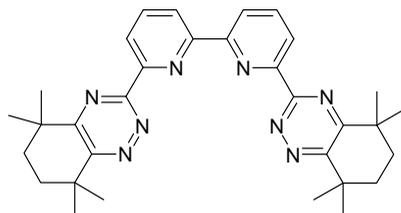
When dealing with a two-phase system like the Chalmers concept it is also important to see if the two extractants extract independently of each other. If so, then a future computer modelling of the system would be much simplified.

How the Chalmers novel GANEX extraction process meets the demands listed above is presented below.

### **Stability towards strong acid**

TBP is a chemical that has been used within the PUREX process for a long time and that has been very well investigated. It has therefore long been known and documented that TBP, to a very small degree, is degraded by nitric acid to di-butyl phosphate, mono-butyl phosphate and finally phosphoric acid [8], which however does not affect the extraction in a negative way. Some of the BTBP molecules on the other hand are much more sensitive to acid and would be rapidly degraded under such a harsh environment as 4 M nitric acid but for instance the CyMe<sub>4</sub>-BTBP (Figure 4) has shown a good stability towards nitric acid [9]. This is because the CyMe<sub>4</sub>-BTBP side chains do not provide any opportunity for hydrogen abstraction on the benzylic carbons.

**Figure 4: Molecular structure of CyMe<sub>4</sub>-BTBP**



Due to these extractants' properties, the GANEX system is stable towards strong acid. The diluent, cyclohexanone, has however been observed to be totally miscible with nitric acid of high concentration but since the high amount of TBP in the GANEX organic system prevents it from being miscible with the water phase, this is not a problem.

### **Actinide extraction**

Extractions of the different actinides U, Np, Pu and Am were performed from 4 M HNO<sub>3</sub> with the new GANEX system consisting of 0.01 M of CyMe<sub>4</sub>-BTBP and 30% TBP in cyclohexanone [10].

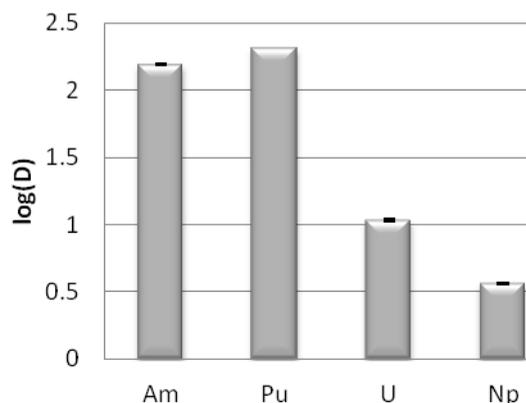
As can be seen in Figure 5, all actinides are readily extracted with the GANEX system from strong acid.

### **An/Ln separation**

Extractions of several lanthanides (La, Nd, Ce, Sm, Eu) were performed with the system to confirm that it could separate them from the actinides as proposed. All lanthanides except Eu were used in the form of inactive salts in as realistic concentrations as possible in the extraction experiments. This means concentrations close to those that can be found in dissolved spent fuel. The europium was however added as <sup>152</sup>Eu in trace amounts [10].

The experiments rendered separation factors between the actinides and the lanthanides that looked very promising even when comparing to the lanthanide that is extracted the most (Eu). When comparing the extraction of the actinides to the extraction of the lanthanide that is the most abundant in spent fuel (Nd) the result looks even better (Table 1). Np is the actinide which has the lowest SF towards the lanthanides but it is still sufficiently high.

**Figure 5: Extraction of actinides ( $^{241}\text{Am}$ ,  $^{238}\text{Pu}$ ,  $^{235}\text{U}$ ,  $^{237}\text{Np}$ ) from 4 M  $\text{HNO}_3$  with 0.01 M  $\text{CyMe}_4\text{-BTBP}$  and 30% TBP in cyclohexanone**



**Table 1: Separation factors for the actinides (U, Np, Pu, Am) and two different lanthanides (Nd, Eu) after extraction from 4 M  $\text{HNO}_3$  with 0.01 M BTBP- $\text{CyMe}_4$  + 30% TBP in cyclohexanone**

Elements	Separation factor
Am/Eu	160
Pu/Eu	210
U/Eu	11
Np/Eu	3.5
Am/Nd	1 700
Pu/Nd	2 300
U/Nd	120
Np/Nd	38

### **Radiolytic stability**

TBP has long been used in the PUREX process where the conditions are more or less the same as they would be in a future GANEX process. The radiolytic degradation of TBP is therefore well known and was not considered to be a problem to the Chalmers concept.

The radiolytic behaviour of the other extractant,  $\text{CyMe}_4\text{-BTBP}$  is however a little bit more uncertain. It has been shown that the molecule can be degraded by  $\gamma$ -radiation [11] but it has also been shown for other BTBP that the degradation is dependent on the diluents as well as the dose rate [12]. Due to this, further investigations of the radiolytic behaviour of the GANEX system are necessary even though preliminary results look promising.

### **Fission and corrosion/activation product extraction**

When extracting several corrosion/activation products (Fe, Ni, Cr, Co, Mn) and fission products (Rb, Sr, Y, Zr, Mo, Ru, Rh, Pd, Ag, Cd, Sb, Te, Cs, Ba) with the GANEX system it was found that a few of the metals were undesirably extracted [10]. Three metals were identified as the most troublesome (Pd, Zr and Mo). This is not, however, an insurmountable problem and there are different ways to approach it. One way is to insert an extraction step before the actual GANEX process during which the troublesome elements are extracted. Another approach would be to add water-soluble complexants to the aqueous phase, hindering the extraction of the unwanted elements in the GANEX process. Previously oxalic acid has been used in i.e. the DIAMEX process as a water soluble complexing agent to stop the extraction of troublesome fission products like zirconium and molybdenum [13]. This is however not applicable in the GANEX process since it is known that oxalic acid precipitates plutonium.

There are some molecules that look promising for the prevention of fission product extraction. One of these is dioctyl sulphide that together with kerosene could be used in a pre-extraction step for Pd [14]. Another one is methionine that has been known to form complexes with Pt and hence could be used as a water-soluble complexing agent for Pd [15]. A third molecule is mannitol, which has been reported to form Mo complexes and therefore could be used to prevent the extraction of this and other metals in the GANEX process [16].

### **Metal loading conditions**

An extraction experiment has been performed where the actinides ( $^{235}\text{U}$ ,  $^{237}\text{Np}$ ,  $^{238}\text{Pu}$  and  $^{241}\text{Am}$ ) and one lanthanide ( $^{152}\text{Eu}$ ) have been added in trace amounts to a 4 M nitric acid water phase loaded with fission products. The metals present were Rb, Sr, Y, Zr, Mo, Rh, Pd, Ag, Cd, Sb, Cs, Ba, La, Ce, Nd, Sm and Te in realistic concentrations [13, 17, 18] rendering a total concentration of over 9 000 ppm [10].

The distribution ratios were somewhat lowered for Am and Pu compared to the case without metal loading but the D for both U and Np stayed the same. Since the D for Eu was decreased this actually rendered an average increase in separation factor (Table 2).

**Table 2: An/Ln separation factors for extraction with 0.01 M CyMe<sub>4</sub>-BTBP and 30% TBP in cyclohexanone from metal loaded (>9 000 ppm) 4 M HNO<sub>3</sub>**

Elements	Separation factor
Am/Eu	101
Pu/Eu	363
U/Eu	91
Np/Eu	28

### **Conclusion**

With the support of several European Union Framework Programmes, much European research within the area of partitioning for transmutation has been conducted over the last 10-15 years. This research has, after many great achievements, in Europe led to the GANEX process. The concept of a GANEX process would bring large benefits industrially by implementing only a very few extraction steps compared to the extraction scheme PUREX-DIAMEX-SANEX-SESAME previously investigated. The novel GANEX extraction process developed at Chalmers University of Technology in Sweden meets many of the basic demands necessary for a viable partitioning process, indicating that GANEX could be a possible future process alternative.

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## Advanced head-end for the treatment of used LWR fuel

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### Abstract

The voloxidation process was initially developed as a dry head-end method for removing tritium from spent uranium–plutonium reactor fuel prior to aqueous processing. Standard voloxidation of  $\text{UO}_2$ -based fuels typically operates at temperatures between 500 and 600°C. This process converts the fuel into a fine  $\text{U}_3\text{O}_8$  powder that can be easily separated from the cladding and completely releases tritium as tritiated water.

A voloxidation process is being developed using these novel oxidising atmospheres: nitrogen dioxide, ozone and mixtures of the two. Un-irradiated pressurised water reactor uranium pellets and simulated fuel containing caesium iodide were used as surrogates. Treatment at significantly lower temperatures using a mixture of  $\text{NO}_2/\text{O}_2$  or  $\text{NO}_2/\text{O}_3$  produced a fine red powder of  $\text{UO}_3$  and released all the iodine (as  $\text{I}_2$ ) from the simulated fuel. This powder – containing the uranium as  $\text{UO}_3$ , transuranium actinides and non-volatile fission product oxides – is readily soluble in dilute acid.

For subsequent aqueous-based separations, these potential improvements can eliminate the prolonged dissolution step and mitigate the presence of iodine in processes downstream of voloxidation, thereby simplifying off-gas treatment. Both of these advantages should result in cost reductions.

The fine  $\text{UO}_3$  powder separated from the cladding is also an excellent initial product for non-aqueous processing methods such as electrochemical separation in molten salts; halide volatility; DUPIC-like processes; supercritical  $\text{CO}_2$  dissolution; aqueous alkaline separations; and hybrid processes (for example, dry chlorination followed by aqueous washing).

## Introduction

The standard voloxidation processing of sheared fuel pieces pulverises the fuel while releasing tritium. The oxidation rate is temperature-dependent and increases with increasing temperature. The increased reaction rate reduces the processing time for batch operations and the residence time for continuous ones and greatly influences the size of the processing equipment required. Although higher operating temperatures are desirable from the perspective of time, changes in fuel dissolution with increasing temperature (i.e. plutonium solubility) need to be considered for optimisation of the process.

The  $\text{UO}_2$  reacts with oxygen to form  $\text{U}_3\text{O}_8$ , which results in a restructuring of the crystallite accompanied by particle crumbling. Comminution increases the available surface area for reaction and releases the fuel from the cladding segments. Tritium, which may be present in the fuel in elemental form, diffuses to the surface of the particle, where it reacts with oxygen to form water, which then enters the gas stream. A fraction of the tritium is retained with the cladding as zirconium hydride ( $\text{ZrT}_x$ ).

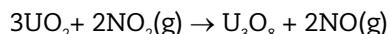
Kilogram-scale tests performed with actual commercial light-water-reactor (LWR) spent fuel ranging from low burn-up and long cooling time to very high burn-up and short cooling time indicated recoveries over 99%. Acid washing of the hulls extended the recovery to 99.9% [1, 2].

Iodine is chemically bonded with caesium and possibly other species; for this reason, only trace amounts of iodine (1% or less) are released in standard voloxidation [3, 4]. The evolution of semi-volatiles at 480°C includes less than about 0.2% of the  $^{106}\text{Ru}$ ,  $^{125}\text{Sb}$  and  $^{134-137}\text{Cs}$ . Higher temperatures increase the fraction of volatiles and semi-volatiles evolved.

## $\text{NO}_2$ voloxidation

Dinitrogen tetroxide ( $\text{N}_2\text{O}_4$ ) is a clear diamagnetic liquid (blue tint) with a boiling point of 21.1°C and a melting point of -9.3°C. It readily dissociates to  $\text{NO}_2$ , which is brown and paramagnetic (i.e. 16% dissociated at 21.1°C, 50% at 40°C and 99% at 135°C). Nitrogen dioxide ( $\text{NO}_2$ ) is a strong oxidiser that is more reactive than  $\text{O}_2$ . Stainless steel and nickel alloys are compatible with  $\text{NO}_2$ , while copper-based alloys are not. Also compatible are fluorinated polymers, such as Teflon; elastomers, such as Kalrez; glass; and quartz.

High-quality pressurised water reactor (PWR) pellets made with natural uranium were used to simulate spent nuclear fuel. Various tests were conducted to evaluate processing at several temperatures using pure  $\text{NO}_2$  and  $\text{NO}_2$ -oxygen mixtures. Initially  $\text{U}_3\text{O}_8$  is formed:



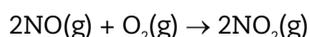
The tests showed the oxidation is faster at significantly lower temperatures than when using air or oxygen at temperatures greater than 450°C. Kinetic studies using a specially designed thermogravimetric system are planned to accurately determine the reaction rates under varied conditions.

The  $\text{U}_3\text{O}_8$  powder readily and completely converts into reddish-brown  $\text{UO}_3$  during cooling:



The resulting  $\text{UO}_3$  products dissolve in a few minutes using warm  $\text{HNO}_3$  at a concentration of 0.25 M or higher, without evolution of  $\text{NO}_x$  gases. This finding is in significant contrast to that for the dissolution of  $\text{UO}_2$  and  $\text{U}_3\text{O}_8$ .

The  $\text{NO}(\text{g})$  by-product readily reacts with  $\text{O}_2$  during cooling to regenerate  $\text{NO}_2$ :



Overall, the process can be autocatalytic, with gaseous  $\text{NO}_2$  being recycled while  $\text{O}_2$  is consumed.

When cooled to near room temperature and treated with pure NO<sub>2</sub> (N<sub>2</sub>O<sub>4</sub>) gas or liquid, the fine red UO<sub>3</sub> powder produces metal nitrates, for example:



Nitrogen dioxide will readily react with O<sub>3</sub> to form N<sub>2</sub>O<sub>5</sub> and possibly NO<sub>3</sub> as an intermediate species. Dinitrogen pentoxide (N<sub>2</sub>O<sub>5</sub>) is a colourless volatile solid at room temperature with a very high vapour pressure. Nitrogen trioxide (NO<sub>3</sub>) is a colourless gas. Voloxidation tests on high-quality surrogate PWR fuel pellets made with natural UO<sub>2</sub> were conducted by mixing gas streams of NO<sub>2</sub> (N<sub>2</sub>O<sub>4</sub>) and a mixture of O<sub>3</sub>/O<sub>2</sub> produced using a commercial ozone generator. The tests showed that the oxidation of the pellets into a fine red UO<sub>3</sub> powder was significantly faster than with NO<sub>2</sub> alone. This finding represents a significant potential improvement, because voloxidation with NO<sub>2</sub> alone is considerably faster than the reactions in air or oxygen at much lower temperatures. Stainless steel components of the system were unaffected.

### Iodine removal

Preliminary tests flowing NO<sub>2</sub> over CsI powder were conducted over a finely dispersed mixture of CsI (5%) in U<sub>3</sub>O<sub>8</sub> powder obtained by voloxidation of UO<sub>2</sub> pellets. On first contact of CsI with NO<sub>2</sub>, the surface becomes black. A sublimate starts to be generated at 50 to 75°C. Two main deposits are collected. One consists of the typical small dark crystals associated with elemental I<sub>2</sub>, and the other consists of shiny grey needles that decompose over time to a black deposit. Most likely, this deposit is unstable nitrosyl iodide (INO), which decomposes to NO and I<sub>2</sub>. The sublimation appears to be complete when the temperature reaches about 130°C. Processing of the CsI/U<sub>3</sub>O<sub>8</sub> mixture results in a final product of red powder, previously identified as UO<sub>3</sub>. When processing CsI alone, the product is a multi-coloured residue, most likely a mixture of caesium oxides (CsO<sub>x</sub>) and CsNO<sub>3</sub>.

The analysis of residual iodine in a large excess of uranium appears to be very difficult. Inductively coupled plasma/mass spectroscopy analyses proved unsuccessful, probably because of the interference of UO<sup>+2</sup> species generated in the plasma. The mass-to-charge ratio for both I<sup>+</sup> and UO<sup>+2</sup> is 127.

An alternative method was then designed for residual iodine analysis. The test was conducted using a 1 g sample of U<sub>3</sub>O<sub>8</sub> that was impregnated with a solution containing 10 mg of non-radioactive CsI spiked with 27 µCi of <sup>125</sup>I. The sample was counted at 35 keV before and after treatments. The sample was contained in a small alumina boat and treated with a gas mixture of NO<sub>2</sub>/O<sub>3</sub> at 325°C for 2 h, producing a bright red UO<sub>3</sub>. The post-treatment sample was counted again, and the activity at 35 keV was 5.7% of the original. A second treatment was conducted using a small rotary calciner for an additional 2 h. The treated sample was counted again, and the residual activity was 1.2% of the original. This test confirmed the efficient iodine removal by NO<sub>2</sub>.

### Summary

A novel dry head-end process is being developed. The main goals are the separation of cladding from fuel and the upfront removal of problematic fission products while facilitating downstream processing. Cold tests using surrogate pellets and tests on a scale of tens of milligrams using actual used fuel are very promising. The initial tests showed that the process can be performed at significantly lower temperatures than those for standard voloxidation by using a mixture of NO<sub>2</sub>/O<sub>2</sub> or NO<sub>2</sub>/O<sub>3</sub> to produce a fine red powder of UO<sub>3</sub> and to release all the tritium and iodine. This powder – containing the uranium as UO<sub>3</sub>, transuranium actinides and non-volatile fission product oxides – is readily soluble in dilute acids. Alternatively, the tests showed that the UO<sub>3</sub> powder can be contacted with NO<sub>2</sub> (N<sub>2</sub>O<sub>4</sub>) gas or liquid at room temperature to produce metal nitrates.

The next steps in the development include kinetic studies using a specially designed thermogravimetric system to accurately determine the reaction rates under varied conditions and the processing of actual spent fuel on a scale of grams to tens of grams.

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## Current progress in R&D on pyrochemical partitioning technology in the Czech Republic

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### Abstract

*The Czech R&D programme in the field of partitioning and transmutation is grounded on the molten salt reactor system concept with fluoride-salt-based liquid fuel, the fuel cycle of which is based on pyrochemical fluoride partitioning of spent fuel. Two main fluoride partitioning technologies applicable within the MSR fuel cycle are under development at the Nuclear Research Institute Řež plc. The first technology devoted to the reprocessing of LWR or FR spent fuel and to the primary processing of MSR transuranium fuel is the fluoride volatility method. The second technology under development is an electrochemical separation process from fluoride molten salt media. The electrochemical separation should be mainly used for “on-line” reprocessing of MSR fuel.*

*R&D on the fluoride volatility method is focused on the development and experimental verification of a semi-pilot technology for reprocessing of current and advanced types of oxide spent fuels from LWR or FR. The technology is based on direct fluorination of powdered spent fuel with fluorine gas and on subsequent separation of fluorinated products based on the differences in their volatility. R&D on electroseparation processes from fluoride molten salt media is focused on the development of a suitable electroseparation technique for partitioning of actinides from fission products in the fluoride melt media. The paper summarises the results achieved in the development of pyrochemical partitioning technologies mentioned above and outlines future activities in the Czech P&T programme.*

## Introduction

The Czech R&D programme in the field of partitioning is based on the development of pyrochemical separation methods, which could be used within the closed fuel cycles of advanced future reactors, chiefly of the molten salt reactor system. The molten salt reactor (MSR) represents one of promising advanced reactor types assigned to the Generation IV reactor systems. It can be operated either as a thorium breeder within the  $^{232}\text{Th}$ - $^{233}\text{U}$  fuel cycle or as an actinide burner (transmuter) incinerating transuranium fuel. The anticipated use of pyrochemical technologies for the reprocessing of spent fuel of advanced reactor systems is based on the fact that the current hydrometallurgical PUREX process will not be suitable for processing several advanced types of nuclear fuels, which are either insoluble in nitric acid or they will not allow the use of any organic solvent due to high production of radioactivity and heat.

Czech activities in the development of pyrochemical partitioning technologies devoted to reprocessing of selected advanced fuel types of Generation IV reactor systems are focused on two fluoride technologies – the fluoride volatility method and electrochemical separations from fluoride molten salt media.

R&D concerning the fluoride volatility method is focused on the development and verification of experimental semi-pilot technology for light water or fast reactor spent fuel reprocessing. Moreover the method can be used as the head-end process within the fuel cycle of molten salt reactor working as a plutonium and minor actinide burner (transmuter). The technology is based on the flame fluorination process and a subsequent partitioning of volatile ( $\text{UF}_6$ ) and non-volatile fluorides ( $\text{PuF}_4$ , minor actinides, fission products) by condensation, sorption and distillation.

R&D on electrochemical separation processes from fluoride melt media is aimed towards the final chemical partitioning of selected actinides from fission products (mostly lanthanides), passed either from the fluoride volatility process or to the on-line reprocessing of the circulating fuel in molten salt reactor. The electrochemical separation technology, under development, is based on a combination of an anodic dissolution method and a cathodic deposition method. The carrier molten salt mixtures examined in the study are mainly  $\text{LiF-BeF}_2$ ,  $\text{LiF-NaF-KF}$  and  $\text{LiF-CaF}_2$  eutectics.

## Progress of the fluoride volatility method

The fluoride volatility method (FVM) is based on a separation process, which is rooted in the specific property of uranium, neptunium and plutonium of forming volatile hexafluorides, whereas most fission products (lanthanides) and higher transplutonium elements present in irradiated fuel form non-volatile trifluorides [1].

The original intention of the development of this dry reprocessing method of spent fuel was motivated in the past by the assumed commercial utilisation of fast breeder reactors. Today, the renewed interest in the dry (pyrochemical) reprocessing methods is motivated by requirements concerning the development of advanced fuel cycle technologies devoted to the Generation IV nuclear reactor systems. Among the pyrochemical technologies under present development, the Fluoride Volatility Method is almost the only one not based on the use of molten salt techniques [2].

The reprocessing technology based on the fluoride volatility method consists of the following main operations:

- fluorination of the powdered fuel (the purpose of this operation is the separation of the uranium component from plutonium, minor actinides and most of the fission products);
- purification of the products obtained.

However before the fluorination of the powdered fuel, the removal of cladding material and subsequent transformation of the fuel into a powdered form of a suitable granulometry has to be done. These two preparatory stages for FVM itself can be realised separately from the FVM. The technology suitable for removing the cladding material is melting in a high-temperature furnace.

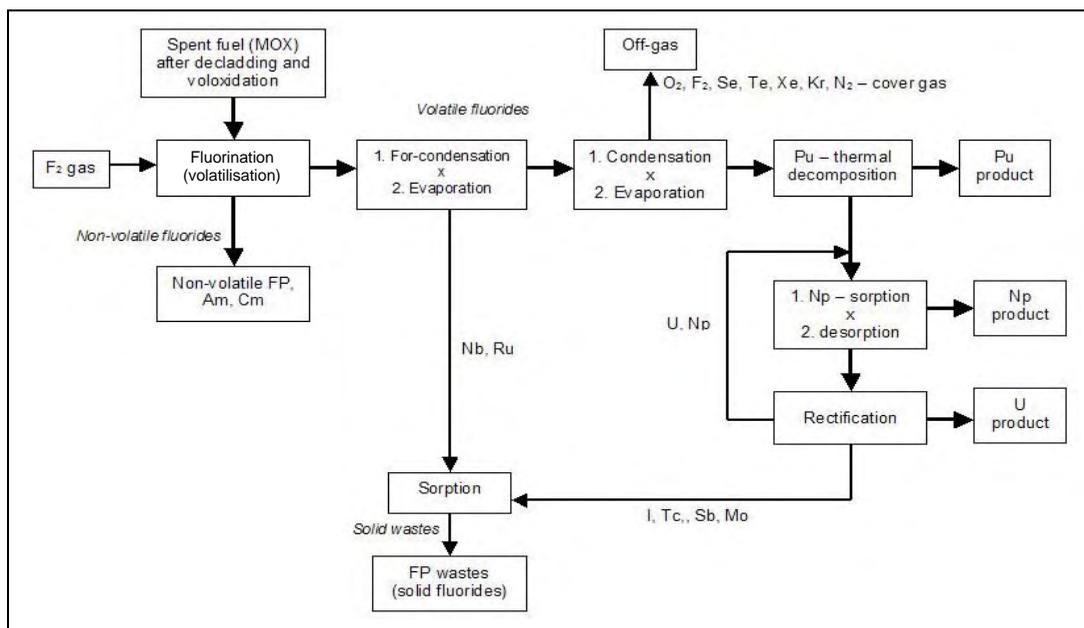
The cladding material of oxide fuel is either zircaloy (LWR) or stainless steel (FR) and both can be fully removed. Transformation of the fuel pellets into a powder is possible either mechanically by grinding or by partial oxidation of  $\text{UO}_2$  into  $\text{U}_3\text{O}_8$ . This chemical process is known as voloxidation. Either flowing air or oxygen at 575 to 650°C is used as an oxidising agent.

Flame fluorination reaction of a spent oxide fuel is a basic unit operation of the whole process. 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 the individual components forming volatile fluorides is generally possible through sorption, condensation or distillation processes. While the fluorination of uranium to the volatile hexavalent form is spontaneous, plutonium hexafluoride is thermally unstable and can be obtained only with a considerable surplus of fluorine gas. The behaviour of neptunium during the flame fluorination varies between uranium and plutonium, however the thermal stability of neptunium hexafluoride is substantially higher than that of plutonium hexafluoride.

Final purification of uranium hexafluorides from  $\text{MoF}_6$ ,  $\text{TcF}_6$ ,  $\text{IF}_5$  and  $\text{SbF}_5$ , which tend to accompany  $\text{UF}_6$  through the system, could be accomplished by a rectification process. Distillation of  $\text{UF}_6$  is usually done in a temperature range from 75 to 90°C at a pressure of about 2 atm in order to produce uranium hexafluorides in liquid form.

The process flow sheet of the FVM, which was designed and calculated, is shown in Figure 1. 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 rectification.

**Figure 1: Process flow sheet of the fluoride volatility method**



For the experimental verification of the technology, the semi-pilot line called FERDA was realised in the radiochemical laboratory of NRI Řež plc. The FERDA line is an experimental technology of batch character with a short-term capacity of 1-3 kg of powdered fuel per hour. The maximum capacity of volatile fluorides ( $\text{UF}_6$ ) accumulated in the technology is about 7 kg. The following technological apparatuses were designed and manufactured for the FERDA line:

- scroll feeders for powdered spent fuel with a capacity of 1-3 kg fuel/hour;
- flame fluorination reactors;

- condensers-evaporators for condensation and evaporation of volatile fluorides;
- sorption columns;
- distillation column for the rectification of a  $UF_6$ - $MoF_6$ - $TcF_6$ - $IF_5$ - $SbF_5$  mixtures.

The current experimental R&D programme is focused mainly on the technological verification of individual unit operations, apparatuses and material research and on the process control. The main experimental effort is focused on the mastering of the fluorination process and on the elimination of technology bottlenecks. After the first fluorination tests done with uranium fuel based on  $U_3O_8$  and  $UO_2$ , the follow-up programme has been aimed at 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.) [3, 4].

### Progress in electrochemical separations from fluoride molten salt media

Electrochemical separation processes seem to be promising as regards final transuranium (TRU) fuel processing for MSR-An-burner and for on-line reprocessing technology of MSR. Here the principle of the partitioning is similar both for MSR-An-burner and for MSR-Th-breeder.

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 rates and their character in the reactor have to be compensated for 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  ${}^7LiF$ - $BeF_2$  remains the basic carrier salt candidate for the MSR primary (fuel) circuit [5].

The main objective of experimental activities in the area of R&D on electrochemical separation technology has been to draft out the separation possibilities of the selected actinides (uranium, thorium) and fission products (lanthanides) in selected fluoride melt carriers. The cyclic voltammetry method was used for studying the basic electrochemical properties [6].

The first step was the choice of fluoride melts suitable for electroseparation. The chosen melt should meet some basic characteristics – low melting point, high solubility of the studied compounds, high electrochemical stability and appropriate physical properties (electrical conductivity, viscosity, etc.). Unfortunately no melt fulfilling all the requirements was found. Therefore three candidate melts were selected for further electrochemical separation studies: eutectic mixture of  $LiF$ - $NaF$ - $KF$  (acronym FLINAK, melting point  $454^\circ C$ , limited electrochemical stability), mixture of  $LiF$ - $BeF_2$  (acronym FLIBE, melting point  $456^\circ C$ , limited electrochemical stability) and eutectic mixture of  $LiF$ - $CaF_2$  (melting point  $766^\circ C$ , good electrochemical stability). A special reference electrode based on the  $Ni/Ni^{2+}$  redox couple was developed to provide reproducible electrochemical measurements in molten fluoride salts.

Selected results of measurements describing the possibilities of individual element separation are listed in Tables 1 and 2 [7, 8].

The general spirit of MSR on-line reprocessing is to maintain the reactor at steady-state conditions by continuous cleaning-up of the primary (fuel) circuit salt. This 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. A conceptual flow sheet of a MSR-An-burner (Molten Salt Transmutation Reactor – MSTR) on-line reprocessing technology is drafted in Figure 2.

**Table 1: Evaluated redox potentials and separation possibilities in FLINAK and LiF-CaF<sub>2</sub>**

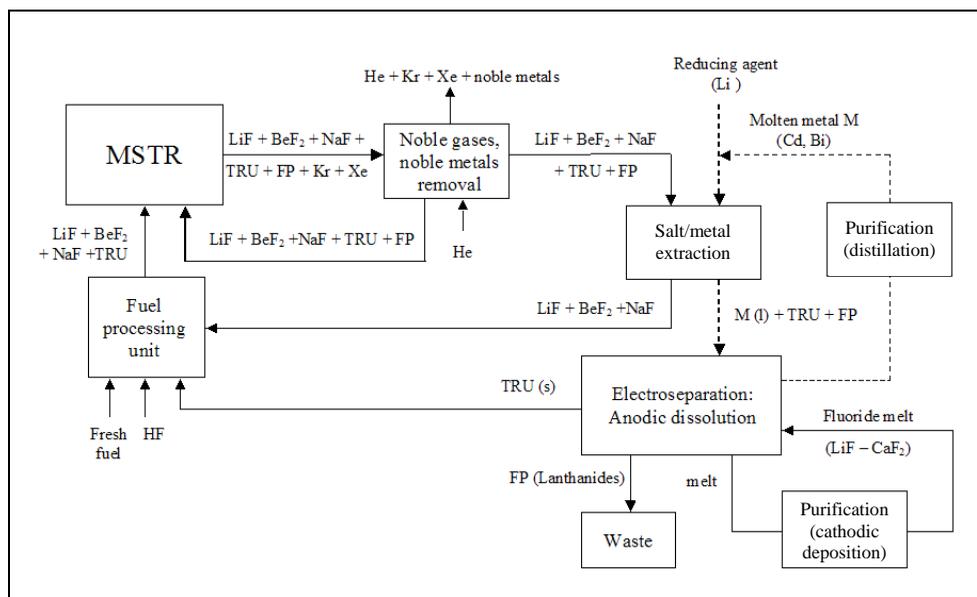
<b>FLINAK</b>	Separated elements	U-Nd	U-Gd	U-Th	U-Eu	Th-Ln	Nd-Gd-Eu
	Potential difference	> 300 mV	> 300 mV	250 mV	200 mV	> 50 mV	> 100 mV
	Separation possibilities	Yes	Yes	Yes	Limit value	N/A	N/A
<b>LiF-CaF<sub>2</sub></b>	Separated elements	U-Nd	U-Gd	U-Eu	Nd-Gd	Nd-Eu	Gd-Eu
	Potential difference	150 mV	250 mV	> 450 mV	100 mV	> 300 mV	> 200 mV
	Separation possibilities	No	Yes	Yes	No	Yes	Yes

**Table 2: Evaluated redox potentials and separation possibilities in FLIBE**

Melt FLIBE		E [V] in FLIBE vs. Ni/Ni <sup>2+</sup>	
Studied reaction	Mechanism	Potential with/without separator	
Melt decomposition on Mo		-1.50	
Uranium reduction on Mo	Two-step	-0.90	-1.40
Melt decomposition on Ni		-1.20	
Behaviour of NdF <sub>3</sub> , GdF <sub>3</sub> , LaF <sub>3</sub> , PrF <sub>3</sub> on Mo	–	Not possible to separate	
ThF <sub>4</sub> on Mo, Ni	–	Not possible to separate	

**Figure 2: Conceptual flow sheet of MSR-An-burner (MSTR) on-line reprocessing technology**

TRU – transuranium elements, FP – fission products

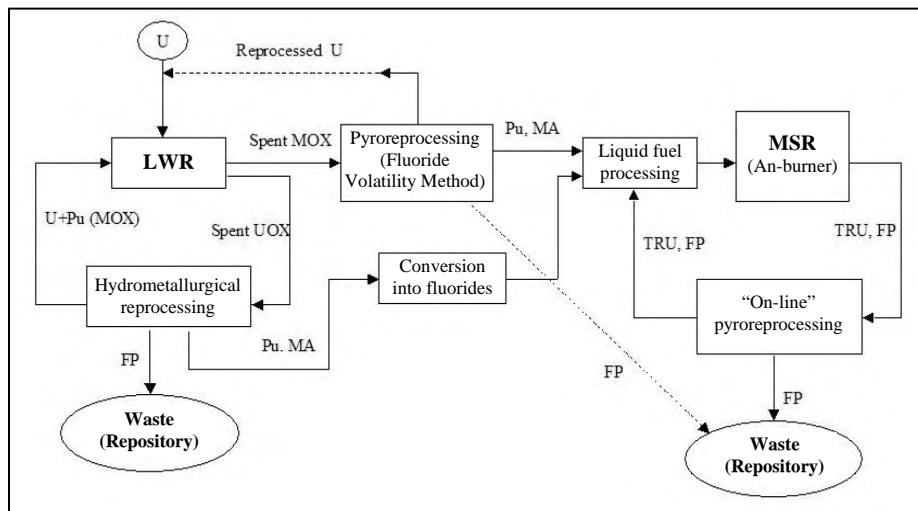


The reprocessing technology drafted in the figure is based on primary non-selective molten-salt/liquid metal reductive extraction and subsequent selective electrochemical separation processes. Li and molten Bi or Cd is proposed to be used as reduction and extraction agents, respectively.

## Conclusion

The fluoride partitioning technologies under development in the Czech Republic can be used mostly within the fuel cycle of molten salt reactor systems. The emplacement of the technologies is shown in Figure 3.

**Figure 3: P&T concept – double-strata strategy with MSR in second stratum**



However the future deployment of molten salt reactor system could also permit a fundamentally different approach to the solution of the transuranium high active waste problem. The new approach could be based on the introduction of the thorium-uranium fuel cycle with the minimised production of transuranium products. The molten salt reactors working within the thorium – uranium fuel cycle in combination with fluoride pyrochemical partitioning technologies could be the promising way how to nearly eliminate the current problem with incineration of long-lived actinides produced by present nuclear reactor systems.

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## Pyropartitioning experiment to recover TRU from high-level liquid waste

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### Abstract

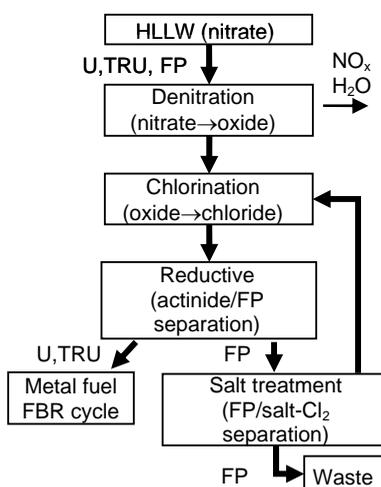
For recovering minor actinide elements (MA) from high-level liquid waste (HLLW) generated by PUREX reprocessing, a pyropartitioning process is being developed. The pyropartitioning process consists of a denitration step which converts various elements in HLLW into oxides by calcination in air, a chlorination step which converts the denitrated material into chlorides by using chlorine gas and carbon, and a reductive-extraction step which separates actinide elements from fission products (FP) in a molten salt/liquid metal system. The recovered actinide elements are expected to be introduced to metal fuel fast reactors for transmutation. The feasibility of the pyropartitioning process was demonstrated using uranium and un-irradiated transuranium elements (TRU). As the next step of the pyropartitioning development, it was necessary to recover TRU from real HLLW and confirm the separability of actinide elements from FP. In the present study, approximately 520 g of HLLW was prepared mainly from the raffinate of PUREX reprocessing of MOX fuel irradiated in a PWR. The HLLW contained 8 400 µg/g of uranium, 600 µg/g of TRU, and 2 000 µg/g of FP including 870 µg/g of rare-earth FP. At the denitration step, the HLLW was calcined at 500°C under air flow. The mass of the denitration product agreed well with the theoretical value estimated from the initial HLLW composition. At the chlorination step, the denitration product mixed with LiCl-KCl eutectic molten salt was chlorinated using pure chlorine gas at 650°C. The analytical results indicated that almost all of actinide elements, rare-earth FP, alkaline-earth FP and alkaline FP remained in the chlorination product salt in their chloride forms. No actinide element was detected in the evaporated part during the denitration and chlorination steps. At the reductive-extraction step, a part of the chlorination product was heated to 500°C together with pure cadmium metal under argon atmosphere. After both salt and cadmium phases were melted, Cd-Li alloy reductant was added to the melts step by step. Finally, almost 100% of each TRU in the initial HLLW was recovered in the liquid cadmium phase, suggesting complete reactions and negligible mass loss in the denitration, chlorination and reductive-extraction steps. The separation behaviours of actinide elements from rare-earth FP observed in the present study were quite similar to those in previous experiments using un-irradiated material. Hence, the pyropartitioning process was successfully verified.

## Introduction

The present PUREX reprocessing of the spent LWR fuel generates high-level liquid waste (HLLW) which will be disposed in a geological repository as a vitrified high-level waste. The vitrified high-level waste contains not only fission products (FP) but also minor actinide elements (MA) such as neptunium, americium and curium. Since MA contain nuclides with quite long half-lives, the recovery of MA from the HLLW would be effective for reducing the toxicity of high-level waste in the future. Additionally, MA also contain heat-generating nuclides such as  $^{241}\text{Am}$ . Therefore, the recovery of MA from the HLLW is also effective for reducing the heat generation of the high-level waste. Since the amount of high-level waste which can be disposed in a geological repository is controlled by heat generation of the high-level waste, the recovery of MA from the HLLW would also be effective to increase the amount of high-level waste that can be disposed of in a repository site. Especially in the cases of processing LWR-MOX fuel and FBR-MOX fuel, the heat generation in the high-level waste caused by the MA is much more than in the case of reprocessing conventional LWR- $\text{UO}_2$  fuel [1]. Therefore, separation of MA from the HLLW will be quite effective for prolonging the repository site's lifetime.

For the recovery of MA from HLLW, the Central Research Institute of Electric Power Industry (CRIEPI) has been developing a pyropartitioning process [2, 3]. A schematic of the pyropartitioning process is shown in Figure 1. In the denitration step, most of the elements in the HLLW, which mainly exist as nitrate forms, are converted to their oxide forms by calcination under air. In the succeeding chlorination step, the elements in the denitrated material are converted to their chlorides in a molten  $\text{LiCl-KCl}$  eutectic salt bath using chlorine gas and carbon. Once elements are converted to their chlorides, the pyrometallurgical methods such as electrolysis in molten salt and reductive-extraction in molten salt/liquid metal system can be adopted for the recovery and separation of actinide elements. The actinide elements are removed from the molten salt as their metallic forms leaving active FP, i.e. rare-earth FP, alkaline-earth FP and alkaline FP, in the molten salt. The recovered actinide elements are used for the fabrication of metal fuel, which mainly consists of U-Pu-Zr alloy. The MA in the metal fuel are transmuted to shorter life nuclides in metal fuel fast reactors [4]. The FP remaining in the molten salt are processed for stabilisation by electrolysis using liquid lead cathode to produce vitrified high-level waste or absorption on zeolite to produce glass-bonded sodalite high-level waste [5-7].

**Figure 1: Schematic of pyropartitioning process**



The pyropartitioning process combined with metal fuel fast reactors has the following merits compared to other partitioning process based on the aqueous process combined with oxide fuel fast reactors:

- It is possible to recover uranium, plutonium and MA together while separating actinide elements from rare-earth FP.
- The solvents used in the pyropartitioning process, i.e. molten salt and liquid metal, are highly radiation resistant and therefore can be used for handling short-cooled and highly concentrated waste with negligible degradation. And generation of secondary waste is less.
- The spent fuel after transmutation can be recycled using the pyroprocess, which is economic even at a small facility such as the processing capacity of 50 tonnes of spent fuel per year.

The targets of the pyropartitioning process are set as follows [3]:

- Recovery ratio of each TRU in the HLLW is more than 99%.
- The total mass of the rare-earth FP in the recovered product is less than that of TRU.

Since the mass of rare-earth FP in HLLW is estimated to be 10 times more than that of the TRU, the second target of the pyropartitioning process corresponds to the decontamination factor of rare-earth FP to be more than 10.

Concerning the development of the pyropartitioning process, experiments using un-irradiated materials such as uranium, TRU and simulating FP were performed. Thermodynamic data including uranium and TRU for electrolysis and reductive-extraction were obtained to assess the process feasibility [8-11]. Through a series of experiments on denitration and chlorination, the behaviours of uranium and FP elements in these steps were investigated [12]. The separation of uranium and TRU from rare-earth elements was also demonstrated by means of the reductive-extraction method [13, 14].

For the development of the pyropartitioning process, a demonstration experiment using real HLLW was necessary to reveal the following unknown matters:

- How do TRU and technetium behave during the denitration and chlorination steps?
- Is there any difference in the separation behaviour between real HLLW and simulated material to be used as the starting material?

In the present study, a hot cell experiment to recover TRU from real HLLW through denitration, chlorination and reductive-extraction was performed in order to demonstrate the pyropartitioning process and to reveal the unknown matters. This study was performed at Institute for Transuranium Elements (ITU) in Germany under a joint research programme between CRIEPI and ITU.

## Experimental

### **Preparation of HLLW**

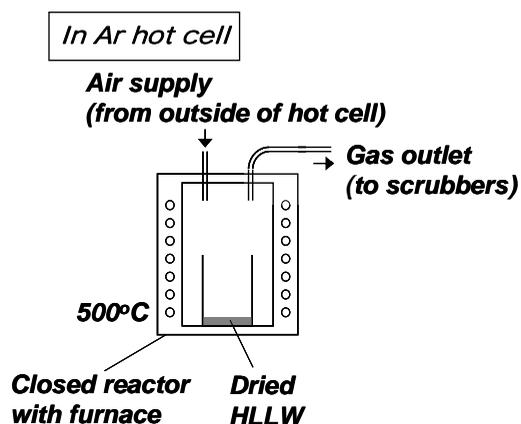
In the present study, raffinate from PUREX reprocessing of a MOX fuel irradiated in a PWR was mainly used. However, the concentrations of neptunium and plutonium in the raffinate were too low to study their behaviours because some fractions of them were already removed during the reprocessing. Therefore, two kinds of solutions containing neptunium and plutonium were added to the raffinate. The prepared HLLW contained roughly 8 400 µg/g of uranium, 600 µg/g of TRU and 2 000 µg/g of FP including 870 µg/g of rare-earth FP. Sodium from the solvent washing and corrosion product elements such as iron were not added to the solution. For the present study, about 520 g of the HLLW was used.

### **Denitration of HLLW**

The entire denitration step was performed in an air atmosphere hot cell. The HLLW solution was firstly heated to 90-120°C in a glass flask for concentration and then dried in a stainless steel crucible by heating to 100-140°C on a hot plate. Afterwards, the dried material in the stainless

steel crucible was calcined at around 500°C as shown in Figure 2. During the calcination, the reactor was purged by air. The exhausted gas from the reactor was lead to a scrubber filled with HNO<sub>3</sub> solution to collect the volatile FP and then lead to another scrubber filled with NaOH solution to capture NO<sub>x</sub> gas formed during the calcination.

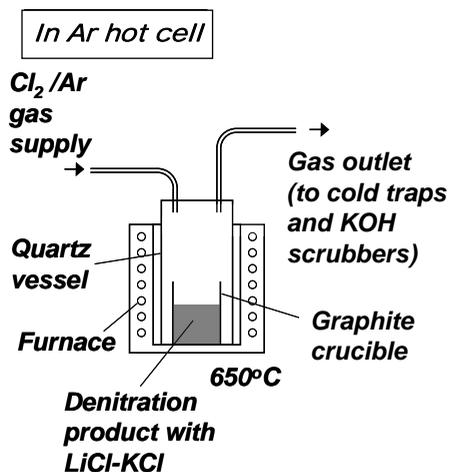
Figure 2: Schematic of denitration



### Chlorination of denitration product

The entire denitration product was charged in a graphite crucible together with 97.5 g of LiCl-KCl eutectic salt in an argon atmosphere hot cell equipped with a vertical furnace. The oxygen and moisture concentrations in the argon atmosphere were kept around 10-20 ppm, respectively. Since the LiCl-KCl eutectic salt purchased from APL Engineered Materials Inc. had a purity of 99.99% and was kept in ampoules under argon, no pre-treatment was conducted to purify the salt before use. As shown in Figure 3, the graphite crucible was placed in a tightly closed quartz vessel, and heated to 650°C. Then, pure chlorine gas was introduced to the vessel in order to chlorinate the denitrated material. The exhausted gas from the vessel was introduced to two cold traps connected in series because some FP were expected to evaporate during the chlorination. Each cold trap contained alumina tube pieces and most of the evaporated material was expected to be collected. The exhausted gas from the cold traps was then lead to two scrubbers containing KOH solutions to capture the un-reacted chlorine gas. The KOH scrubbers might also have worked to collect volatile FP if they were present. Finally, the exhausted gas from the KOH scrubbers was lead to the ventilation system of the facility after monitoring the Cl<sub>2</sub> concentration.

Figure 3: Schematic of chlorination



### Reductive-extraction from chlorination product

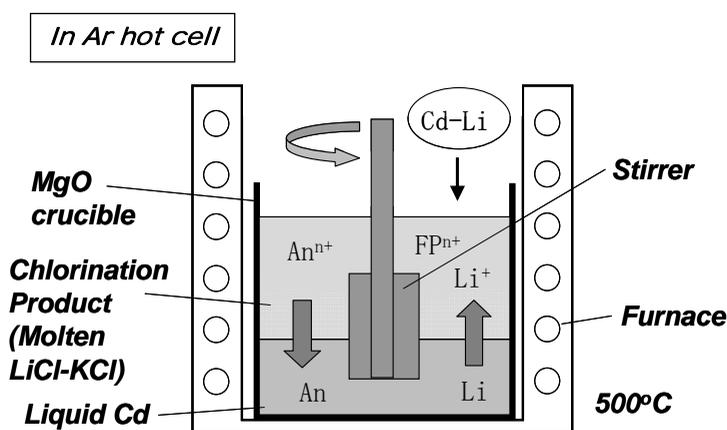
For the following reductive-extraction, 47.3 g of the chlorination product salt was recovered in an MgO crucible. 204.5 g of cadmium metal, which was purchased from Rare Metallic Co. Ltd. and had a purity of 99.9999%, was also loaded in the crucible. After heating to 500°C in the furnace, samples were taken from both molten salt and liquid cadmium phases. The salt sample was taken by dipping a cold stainless steel rod into the molten salt. The cadmium sample was taken by sucking the liquid cadmium into a narrow glass tube connected to a syringe. After cooling to ambient temperature, the glass tube was broken to recover the solidified cadmium sample.

A schematic of the reductive-extraction is shown in Figure 4. In order to reduce the elements in the salt phase according to the reaction indicated in Eq. (1), Cd-Li alloy reductant containing 0.685 wt.% of lithium was added to the melt step by step.



The Cd-Li alloy was previously prepared by mixing cadmium metal of 99.9999% purity and lithium metal of 99.9% purity, which were purchased from Rare Metallic Co. Ltd.

Figure 4: Schematic of reductive-extraction



The reductive-extraction step was also performed in the same argon atmosphere hot cell as the chlorination. A stainless steel stirrer was used to homogenise the melt after each addition of the reductant. After equilibrium was attained, both molten salt and liquid cadmium samples were taken as previously described. The sequence of “Cd-Li alloy addition”, “stirring” and “taking samples” were repeated 14 times. The theoretical amounts of the Cd-Li reductant necessary to reduce uranium, TRU and rare-earth FP contained in the initial salt phase, which were determined from the chemical analysis of the chlorination product sample, were 28.2 g, 2.0 g and 4.0 g, respectively. In order to reduce these elements completely, totally 45.1 g of the Cd-Li alloy was added in the present study, which was 45% more than the sum of the above theoretical values, i.e. 34.2 g.

### Analysis

Sample preparation for analysis was performed in an air atmosphere hot cell. Liquid samples such as scrubber solutions were diluted with nitric acid. In order to collect the evaporated material during denitration and chlorination, the inside of the connecting tubes and the cold traps were washed by nitric acid and diluted. Salt samples taken during the chlorination and the reductive-extraction were once dissolved in water and then diluted with nitric acid. The cadmium samples taken at the reductive-extraction were once washed by water in order to remove adhering salt and then dissolved in nitric acid. The diluted solutions were analysed by an Inductively Coupled Plasma-Mass Spectrometry (ICP-MS); ELEMENT2 manufactured by Thermo Scientific in Germany.

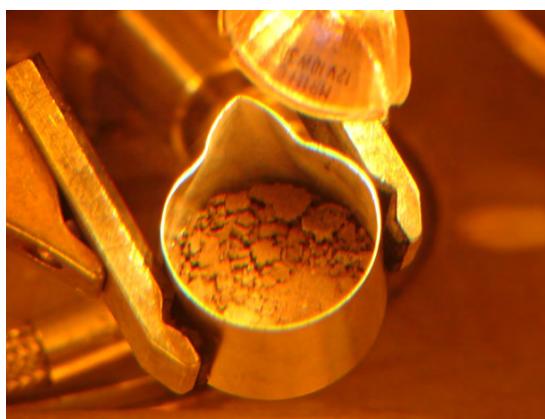
## Results

### *Denitration of HLLW*

After the first calcination for 20.5 hours, the vessel was weighed. Then, the material was subject to the second calcination for 67.5 hours. Since the weight loss during the second calcination was only 27 mg, no more calcination was performed.

The final weight of the denitrated material was 7.41 g. Almost all of the denitrated material, 7.32 g, was successfully recovered from the crucible, as shown in Figure 5. No damage or reaction was observed on the inside surface of the stainless steel crucible. The mass of the calcined material agreed with its theoretical value of 6.91 g, which was estimated from the initial HLLW composition. According to the ICP-MS analysis of scrubbers and connecting tube washing solutions, only 0.3% of ruthenium in the initial HLLW was detected as the evaporated material.

**Figure 5: Recovered material after denitration**



### *Chlorination of denitration product*

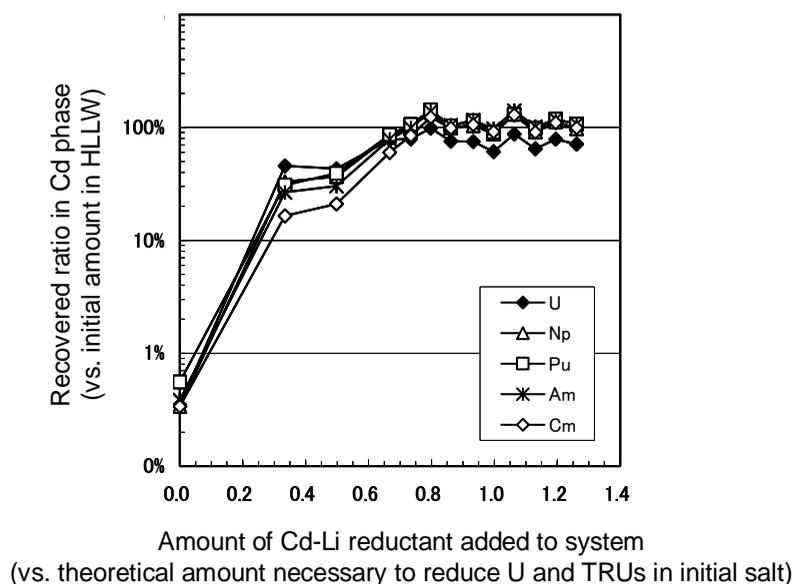
After the introduction of the chlorine gas, some brown deposits was observed in the gas outlet tube. The section of the product salt after breaking the graphite crucible seemed almost uniform and no precipitate indicating un-reacted denitrated materials was observed.

The chemical analysis results of the chlorination samples indicated that almost all of the actinide elements, rare-earth FP, alkaline-earth FP and alkaline FP remained in the chlorinated product salt, probably as their chloride forms. Therefore, no mass loss of the actinide elements was detected during denitration and chlorination. Though very little amount of ruthenium was detected as the evaporated material at the denitration step, most of technetium and noble metal fission products remained in the chlorination product. On the contrary, some fission products such as zirconium and molybdenum evaporated during the chlorination as expected. According to the above considerations it can be deduced that the chlorination was successful.

### *Reductive-extraction from chlorinated product*

#### *Recovery of actinide elements from salt phase*

The concentrations of actinide elements in the salt phase were gradually reduced by the addition of Cd-Li alloy and no actinide elements were consequently detected in the salt. As shown in Figure 6, TRU were almost completely recovered in the cadmium phase. Therefore, complete reactions and negligible mass loss in denitration, chlorination and reductive-extraction steps were observed. The slightly poor mass balance of uranium is considered to be owing to formation of  $\text{UO}_2$  induced by impurities in the argon atmosphere such as oxygen and moisture.

**Figure 6: Recovered ratios of uranium and TRU in cadmium phases**

#### *Behaviours of FP during reductive-extraction*

The trivalent rare-earth FP such as lanthanum and neodymium were reduced from the salt phase just after the reduction of actinide elements started, and then reduction of the divalent rare-earth FP such as europium started. Alkaline FP and alkaline-earth FP, such as caesium and strontium, were not reduced throughout the reductive-extraction. The concentrations of FP in the cadmium phase increased according to the reduction from the salt phase.

The trivalent rare-earth FP showed similar mass balance profiles to those of TRU and were almost 100% versus the initial HLLW. The mass balance profiles of divalent rare-earth FP were slightly worse when the amount of Cd-Li alloy addition exceeded 30 g, probably because the mass in the cadmium phase was not correctly analysed. The mass balances of alkaline metal FP were also nearly 100% throughout the reductive-extraction. On the contrary, the mass balances of transition metal FP, noble metal FP and chalcogen FP were not stable and poor in general. For molybdenum, zirconium and chalcogen FP, it was probably due to evaporation during the chlorination. For technetium and noble metal FP, the reasons were not clarified. Un-uniform compositions in the chlorination product might be one of the reasons because technetium and noble metal FP are expected to be insoluble in the molten salt phase.

#### *Separation behaviour between actinide elements and rare-earth FP*

The separation factor (SF) in a molten salt/liquid cadmium system is defined using Eq. (2) [8]:

$$SF_{A/B} = \frac{(\text{mole fraction of A in salt}) \cdot (\text{mole fraction of B in Cd})}{(\text{mole fraction of A in Cd}) \cdot (\text{mole fraction of B in salt})} \quad (2)$$

where  $SF_{A/B}$  denotes the separation factor of element A versus element B.

In the case of both elements A and B take the same valence in the salt phase like actinide and most of the rare-earth elements, which take trivalent in the salt, the SF is theoretically constant [8].

In the present study, the distributions of the major nuclides versus  $^{238}\text{U}$  lie on lines with constant slope, as theoretically estimated. As shown in Table 1, the SF of the major nuclides versus  $^{238}\text{U}$  obtained in the present study agreed well with the literature values obtained by

**Table 1: Separation factors of TRU and rare-earth FP versus  $^{238}\text{U}$  in molten LiCl-KCl/Cd system at 500°C**

Nuclide	$^{237}\text{Np}$	$^{239}\text{Pu}$	$^{243}\text{Am}$	$^{244}\text{Cm}$	$^{139}\text{La}$	$^{140}\text{Ce}$	$^{141}\text{Pr}$	$^{143}\text{Nd}$
SF in present study	2.0	1.6	3.1	4.1	113	44	40	44
Literature value	2.12	1.88	3.08	3.93*	131	49	42	45

\* Obtained at 510°C.

un-irradiated materials [8, 9, 11]. This means that the separation behaviour of irradiated material is similar to the un-irradiated material. Since the difference of SF between actinide elements and rare-earth FP is not large enough to attain the partitioning criteria in a single extraction system, a multi-stage counter-current reductive-extraction technology is under development [15].

## Conclusion

A hot cell experiment to recover transuranium elements (TRU) from real high-level liquid waste (HLLW) through denitration, chlorination and reductive-extraction was performed. Through the chlorination of the denitration product, all of the actinide elements were converted to their chloride forms. By the reductive-extraction using the chlorination product, nearly 100% of each TRU contained in the initial HLLW was recovered in the liquid cadmium phase. Thus, complete reaction and negligible mass loss in the denitration, chlorination and reductive-extraction steps were observed. The separation behaviours of actinide elements from fission products (FP) were quite similar to the previous results obtained using un-irradiated material. These results indicate that the predicted reaction in each step will be completed, and that actinide elements will be separated from FP in the actual separation system. Hence, the pyropartitioning process was successfully verified.

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## **Session V**

### **Transmutation Physics and Materials**

***Chairs: E.M. González-Romero, C. Fazio***



## Pb alloys' coolant chemistry and structural materials performance: A review\*

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### Abstract

*Innovative nuclear systems cooled with liquid Pb and Pb-Bi eutectic (LBE) such as e.g. the accelerator-driven transmutation system or the lead fast reactor can be potentially used for the transmutation of high-level nuclear wastes. The safe operation of these systems implies, among other items, a detailed knowledge of the materials' performance in the liquid metal environment and the handling of the liquid metal in terms of soluble and insoluble impurities. The objective of this contribution is to summarise relevant studies conducted recently in Europe on materials selection and characterisation in terms of corrosion and mechanical resistance in the liquid metal. The analysis will be done by taking into account the importance of the HLM chemistry. In particular the oxygen effect will be illustrated and techniques able to adjust and measure the oxygen content in the liquid metal will be described.*

*As far as the selection of the structural materials for the core components, vessel and in-vessel components of liquid-metal-cooled systems, this selection is dictated by several factors such as temperature, mechanical stresses, irradiation damage and corrosion resistance.*

*The corrosiveness of the liquid Pb and LBE has been thoroughly investigated and it has been found that liquid metal chemistry, temperature and flow rate have an important role in the appearance of the corrosion mechanism and in the estimation of the corrosion rate. In particular the oxygen dissolved in the liquid metal, in combination with the different temperature ranges, has the most important effect on the steel corrosion mechanism. Indeed, steels containing elements highly soluble in the liquid metals as, e.g. the class of austenitic steels, are oxidised when the level of oxygen in the liquid reaches adequate potentials and the oxidation rate does not exceed tenths of micrometers per year. However, even under oxidising conditions the same steel suffers severe dissolution corrosion for temperatures above ~450°C. Dissolution attack depends on the temperature and it can reach even hundreds of micrometers of penetration from the surface to the bulk. The consequences of severe dissolution of austenitic steels is a change in the microstructure of this material, since Ni is leached out (due to the highest solubility in the Pb and LBE) and the austenitic structure changes to ferritic until other elements such as Cr and Fe are also dissolved. This mechanism can induce a reduction of the strength, i.e. the load-bearing capability of the steel.*

*On the other hand the class of ferritic/martensitic steels has a different behaviour. Indeed, these steels are oxidised with a considerable rate in the high-temperature range (~500-550°C). A high oxidation rate implies a thick oxide layer which can spall off and allow liquid metal penetration with consequent change of corrosion mechanism from oxidation to dissolution. These events must be avoided, since ferritic/martensitic steels suffer mechanical properties degradation due to the liquid metal. However, a proper oxygen potential control in the liquid metal and for lower temperatures (< 500°C) the oxide layer may act as corrosion protection barrier and the thickness*

\* The full paper being unavailable at the time of publication, only the abstract is included.

*of the scale may stay within acceptable levels. The control, in terms of measurement and setting, of the oxygen potential in the liquid metal can occur with dedicated techniques. Indeed, oxygen potential measurement can be done with electrochemical probes and the adjustment of the oxygen level can be performed through the gas phase or mass exchange devices. Electrochemical probes with different reference electrodes have been developed and it turned out that the Pt/air reference electrode is very reliable for oxygen potential measurement in the high-temperature range, while Me/MeO (Me: Bi, In, etc.) are reference electrodes that can be used for measurement in lower temperature ranges.*

*As stated above, the objective of this contribution is to illustrate the relation between structural materials' performance and liquid metal chemistry and the impact of these two items on design and safety aspects of nuclear transmutation reactors cooled with HLM.*

### **Acknowledgements**

The author wants to acknowledge all partners of the EUROTRANS project and in particular of the DEMETRA Domain, since relevant results summarised here have been published within this project. Moreover, a special acknowledgment goes to the European Commission, which has supported the EUROTRANS project under contract number FI6W 516520.

## Critical analysis of the effective delayed neutron fraction calculation routes by deterministic and Monte Carlo codes

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### Abstract

The studies on accelerator-driven systems (ADS) have renewed interest in the theoretical and computational evaluation of the main integral parameters characterising subcritical systems (e.g. reactivity, effective delayed neutron fraction, mean neutron generation time). In particular, some kinetic parameters, such as the effective delayed neutron fraction, are evaluated in Monte Carlo codes by formulations which do not require the calculation of the adjoint flux. This paper is focused on a theoretical and computational analysis of how the different  $\beta_{\text{eff}}$  definitions are connected, and which are the approximations inherent to the Monte Carlo definition with respect to the standard definition involving weighted integrals. By means of a refined transport computational analysis carried out in a coherent and consistent manner, i.e. using the same deterministic code and neutron data library for the  $\beta_{\text{eff}}$  evaluation in different ways, the theoretical analysis is numerically confirmed. Both theoretical and numerical results confirm the effectiveness of the Monte Carlo  $\beta_{\text{eff}}$  evaluation by the relationship  $1 - (k_p/k)$ , at least in cases where spectral differences between total and prompt fluxes are negligible with respect to the value of the functionals entering the classical  $\beta_{\text{eff}}$  formulation.

## Introduction

The studies on accelerator-driven systems (ADS) have renewed interest in the theoretical and computational evaluation of the main integral parameters characterising subcritical systems (e.g. reactivity, effective delayed neutron fraction, mean neutron generation time [1-3]). In particular, the extensive use of Monte Carlo codes for the analysis of the ADS neutronic behaviour is challenging the deterministic codes, classically used in the past for the analysis of critical systems, as concerns the capability to reproduce Monte Carlo results, usually taken as reference. Nevertheless, some particular parameters, such as the effective delayed neutron fraction, are evaluated in Monte Carlo codes by formulations which do not require the calculation of the adjoint flux. The assessment of the various formulations of the effective delayed neutron fraction is crucial for the system evaluation, since it plays an important role in determining its dynamic characteristics. This paper is focused on a theoretical and computational analysis as to how the different  $\beta_{\text{eff}}$  definitions are connected, and which are the approximations inherent to the Monte Carlo definition with respect to the standard definition involving weighted integrals. Theoretical results show how the Monte Carlo formulation of  $\beta_{\text{eff}}$  may be connected to the classical definition, interpreting the classical one through a reactivity evaluation based on an “improved” first-order approach of perturbation theory. The computational analysis is carried out in a coherent and consistent manner, using the same deterministic code and neutron data library for the  $\beta_{\text{eff}}$  evaluation. A simplified system allows to investigate the features of the various procedures and to use results to obtain a physical insight. The GUINEVERE system is then selected as a relevant test case for ADS technology. The GUINEVERE experience [4, 5], mainly devoted to the issues concerning on-line reactivity monitoring in ADS, is analysed by using a modified layout of the VENUS critical facility located at the Belgium SCK•CEN Mol site, coupling the subcritical core facility to a deuteron accelerator delivering, by a continuous or pulsed beam, 14 MeV neutrons by deuterium-tritium reactions. For the GUINEVERE experience the ERANOS system [6] and neutron data library JEFF-3.1 [7] have been used to perform transport calculations with 49 energy groups for a cylindrical schematisation of the GUINEVERE start-up (at critical) configuration. The analysis of the results allows drawing some conclusions on the merits and the limits of the various formulations.

## Theoretical analysis

### The general case

Let us take as the reference system the following eigenvalue problem:

$$\mathbf{L}\varphi = \omega\mathbf{F}\varphi$$

where  $\mathbf{L}$  is the loss operator,  $\mathbf{F}$  the fission operator and  $\omega = 1/k$ . A perturbation  $\delta\mathbf{F}$  is then introduced in the system. As a consequence, the eigenvalue problem for the perturbed system is:

$$\mathbf{L}\varphi^* = [\omega + \delta\omega][\mathbf{F} + \delta\mathbf{F}]\varphi^* \quad (1)$$

with  $\varphi^*$  as the perturbed flux. Following the exact approach of perturbation theory (PT), i.e. considering the adjoint problem corresponding to the reference system:

$$\mathbf{L}^+\varphi^+ = \omega\mathbf{F}^+\varphi^+$$

we obtain<sup>1</sup> (<...> stands for integration):

$$\frac{\delta\omega}{\omega + \delta\omega} = - \frac{\langle \varphi^+ \delta\mathbf{F} \varphi^* \rangle}{\langle \varphi^+ \mathbf{F} \varphi^* \rangle} \quad (2)$$

1. Or equivalently:  $\frac{\delta\omega}{\omega} = - \frac{\langle \varphi^+ \delta\mathbf{F} \varphi^* \rangle}{\langle \varphi^+ (\mathbf{F} + \delta\mathbf{F}) \varphi^* \rangle}$

If  $\omega + \delta\omega = 1/k^*$  we can write Eq. (2) in terms of  $k$  eigenvalues:

$$1 - \frac{k^*}{k} = - \frac{\langle \varphi^+ \delta \mathbf{F} \varphi^* \rangle}{\langle \varphi^+ \mathbf{F} \varphi^* \rangle} \quad (3)$$

If we write Eq. (1) as:

$$\mathbf{L}[\varphi + \delta\varphi] = [\omega + \delta\omega][\mathbf{F} + \delta\mathbf{F}][\varphi + \delta\varphi]$$

and we follow the first-order PT approach, while retaining the second-order term  $\delta\omega\delta\mathbf{F}\varphi$ , we obtain the “improved” first-order formulation:

$$\frac{\delta\omega}{\omega + \delta\omega} \cong - \frac{\langle \varphi^+ \delta \mathbf{F} \varphi \rangle}{\langle \varphi^+ \mathbf{F} \varphi \rangle}$$

or in terms of  $k$  eigenvalues:

$$1 - \frac{k^*}{k} \cong - \frac{\langle \varphi^+ \delta \mathbf{F} \varphi \rangle}{\langle \varphi^+ \mathbf{F} \varphi \rangle} \quad (4)$$

### The “prompt” perturbation

If the perturbation is  $\delta\mathbf{F} = -\mathbf{F}_d$ , with  $\mathbf{F}_d$  as delayed neutrons fission operator, the perturbed system given in Eq. (1) can be written as:

$$\mathbf{L}\varphi_p = \omega_p[\mathbf{F} - \mathbf{F}_d]\varphi_p$$

with the perturbed flux given by the prompt flux  $\varphi_p$  and  $\omega_p = 1/k_p$ . Then Eq. (3) becomes:

$$1 - \frac{k_p}{k} = \frac{\langle \varphi^+ \mathbf{F}_d \varphi_p \rangle}{\langle \varphi^+ \mathbf{F} \varphi_p \rangle} \quad (5)$$

and Eq. (4):

$$1 - \frac{k_p}{k} \cong \frac{\langle \varphi^+ \mathbf{F}_d \varphi \rangle}{\langle \varphi^+ \mathbf{F} \varphi \rangle} = \beta_{\text{eff}} \quad (6)$$

Thus, the effective delayed neutron fraction  $\beta_{\text{eff}}$  is an “improved” PT first-order formulation of the relationship  $1 - (k_p/k)$ , widely used in Monte Carlo codes as  $\beta_{\text{eff}}$  estimator.<sup>2</sup> To pass from Eq. (5) to Eq. (6) it is sufficient to replace  $\varphi_p$  by  $\varphi$ .

### Standard and Monte Carlo $\beta_{\text{eff}}$ formulations

The effectiveness of delayed neutrons is normally obtained by one of the following two relationships:

$$\beta_{\text{eff}} = \sum_m \sum_i \bar{v}_{d,i}^{(m)} \frac{\langle\langle \varphi^+(\mathbf{r}, E) \chi_{d,i}^{(m)}(E) \langle \Sigma_f^{(m)}(\mathbf{r}, E') \varphi(\mathbf{r}, E') \rangle_{E'} \rangle_{E, \mathbf{r}}}{\sum_m \langle\langle \varphi^+(\mathbf{r}, E) \chi^{(m)}(E) \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \varphi(\mathbf{r}, E') \rangle_{E'} \rangle_{E, \mathbf{r}}} \quad (7)$$

$$\tilde{\beta}_{\text{eff}} = \sum_m \sum_i \beta_i^{(m)} \frac{\langle\langle \varphi^+(\mathbf{r}, E) \chi_{d,i}^{(m)}(E) \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \varphi(\mathbf{r}, E') \rangle_{E'} \rangle_{E, \mathbf{r}}}{\sum_m \langle\langle \varphi^+(\mathbf{r}, E) \chi^{(m)}(E) \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \varphi(\mathbf{r}, E') \rangle_{E'} \rangle_{E, \mathbf{r}}} \quad (8)$$

2. The pure PT first-order formulation provides:  $\frac{\delta\omega}{\omega} = \frac{k}{k_p} - 1 \cong - \frac{\langle \varphi^+ \delta \mathbf{F} \varphi \rangle}{\langle \varphi^+ \mathbf{F} \varphi \rangle} = \frac{\langle \varphi^+ \mathbf{F}_d \varphi \rangle}{\langle \varphi^+ \mathbf{F} \varphi \rangle} = \beta_{\text{eff}}$

where:  $m$  fissile isotope index;

$i$  delayed neutron group index;

$\chi_{d,i}^{(m)}$  delayed neutron spectrum for fissile isotope  $m$ , delayed neutron group  $i$  (basic data);

$\chi^{(m)}$  total neutron spectrum for fissile isotope  $m$ ;

$\bar{v}_{d,i}^{(m)}$  average value of delayed neutrons emitted from fissile isotope  $m$ , delayed neutron group  $i$ , for a given incident neutron spectrum (from basic data);

$v^{(m)}$  total neutrons emitted from fissile isotope  $m$ ;

$\Sigma_f^{(m)}$  macroscopic fission cross-section for fissile isotope  $m$ ;

and, furthermore:

$$\beta_i^{(m)} = \frac{\bar{v}_{d,i}^{(m)}}{\bar{v}_\psi^{(m)}} \quad (9)$$

with  $\bar{v}_\psi^{(m)}$  average value of total neutrons emitted from fissile isotope  $m$  for a given incident neutron spectrum  $\psi$  ( $\psi$  has to be calculated by a cell or system neutron calculation). In Eqs. (7) and (8) the direct flux and the adjoint flux are solutions of the following eigenvalue equations, respectively:

$$\Omega \cdot \nabla \varphi(\mathbf{r}, E) + \Sigma_t(\mathbf{r}, E)\varphi(\mathbf{r}, E) = \frac{1}{k} \sum_m \chi^{(m)}(E) \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \varphi(\mathbf{r}, E') \rangle_{E'} + \langle \Sigma_s(\mathbf{r}, E' \rightarrow E) \varphi(\mathbf{r}, E') \rangle_{E'} \quad (10)$$

$$-\Omega \cdot \nabla \varphi^+(\mathbf{r}, E) + \Sigma_t(\mathbf{r}, E)\varphi^+(\mathbf{r}, E) = \frac{1}{k} \sum_m v^{(m)}(E) \Sigma_f^{(m)}(\mathbf{r}, E) \langle \chi^{(m)}(E') \varphi^+(\mathbf{r}, E') \rangle_{E'} + \langle \Sigma_s(\mathbf{r}, E \rightarrow E') \varphi^+(\mathbf{r}, E') \rangle_{E'} \quad (11)$$

Current Monte Carlo (MC) calculations approximate  $\beta_{\text{eff}}$  by the following formula:

$$\beta_{\text{eff}}^{\text{MC}} = 1 - \frac{k_p}{k} \quad (12)$$

where  $k_p$  is the eigenvalue of the equation:

$$\Omega \cdot \nabla \varphi_p(\mathbf{r}, E) + \Sigma_t(\mathbf{r}, E)\varphi_p(\mathbf{r}, E) = \frac{1}{k_p} \sum_m \left[ \chi^{(m)}(E) - \sum_i \chi_{d,i}^{(m)}(E) \beta_i^{(m)}(\mathbf{r}) \right] \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \varphi_p(\mathbf{r}, E') \rangle_{E'} + \langle \Sigma_s(\mathbf{r}, E' \rightarrow E) \varphi_p(\mathbf{r}, E') \rangle_{E'} \quad (13)$$

Eq. (13) is written in a general form, taking into account the spatial dependence of the delayed emission fraction  $\beta$  on position  $\mathbf{r}$ . In this case  $\beta_i^{(m)}(\mathbf{r})$  is given by:

$$\beta_i^{(m)}(\mathbf{r}) = \frac{\bar{v}_{d,i}^{(m)}}{\left[ \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \varphi_p(\mathbf{r}, E') \rangle_{E'} / \langle \Sigma_f^{(m)}(\mathbf{r}, E') \varphi_p(\mathbf{r}, E') \rangle_{E'} \right]} \quad (14)$$

If we consider the system governed by Eq. (13) as a perturbation of the system governed by Eq. (10), we obtain as exact perturbation Eq. (5), i.e.:

$$1 - \frac{k_p}{k} = \sum_m \sum_i \frac{\langle \langle \varphi^+(\mathbf{r}, E) \chi_{d,i}^{(m)}(E) \beta_i^{(m)}(\mathbf{r}) \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \varphi_p(\mathbf{r}, E') \rangle_{E'} \rangle_{E} \rangle_{\mathbf{r}}}{\sum_m \langle \langle \varphi^+(\mathbf{r}, E) \chi^{(m)}(E) \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \varphi_p(\mathbf{r}, E') \rangle_{E'} \rangle_{E} \rangle_{\mathbf{r}}} \quad (15)$$

with  $\phi^+$  eigenfunction of Eq. (11) and  $\phi_p$  eigenfunction of Eq. (13). When comparing Eq. (15) with Eq. (8) it can be noticed that, besides the replacement of  $\phi$  by  $\phi_p$ , the differences lie in the presence of the terms  $\beta_i^{(m)}(\mathbf{r})$  (under integration) replacing the terms  $\beta_i^{(m)}$ . If we assume in both Eqs. (13) and (15) the following approximation:

$$\beta_i^{(m)}(\mathbf{r}) \approx \beta_i^{(m)} = \frac{\bar{v}_{d,i}^{(m)}}{\bar{v}_\psi^{(m)}} \quad (16)$$

Eq. (13) takes the following form:

$$\begin{aligned} \Omega \cdot \nabla \phi_p(\mathbf{r}, E) + \Sigma_t(\mathbf{r}, E) \phi_p(\mathbf{r}, E) = \\ \frac{1}{\bar{k}_p} \sum_m \left[ \chi^{(m)}(E) - \sum_i \chi_{d,i}^{(m)}(E) \beta_i^{(m)} \right] \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \phi_p(\mathbf{r}, E') \rangle_{E'} + \langle \Sigma_s(\mathbf{r}, E' \rightarrow E) \phi_p(\mathbf{r}, E') \rangle_{E'} \end{aligned} \quad (17)$$

and Eq. (15) becomes:

$$\tilde{\beta}_{\text{eff}}^{\text{MC}} = 1 - \frac{\bar{k}_p}{\bar{k}} = \sum_m \sum_i \beta_i^{(m)} \frac{\langle \langle \phi^+(\mathbf{r}, E) \chi_{d,i}^{(m)}(E) \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \phi_p(\mathbf{r}, E') \rangle_{E'} \rangle_E \rangle_{\mathbf{r}}}{\sum_m \langle \langle \phi^+(\mathbf{r}, E) \chi^{(m)}(E) \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \phi_p(\mathbf{r}, E') \rangle_{E'} \rangle_E \rangle_{\mathbf{r}}} \quad (18)$$

As shown in the previous section, the classical  $\beta_{\text{eff}}$  definition given in Eq. (8) is an “improved” PT first-order approximation of the relationship  $1 - (\bar{k}_p/\bar{k})$  given in Eq. (18). From the point of view of the perturbation approach, leading to a relationship among system integral properties like the eigenvalues and the delayed neutron effectiveness, Eqs. (17) and (18) are intimately coupled, in the sense that the coherent prompt flux  $\phi_p$  to be used in the  $\beta_{\text{eff}}$  definition given by Eq. (18) is the eigenfunction of Eq. (17). We recall that Eq. (17) is an approximation of Eq. (13) owing to the assumption given in Eq. (16). If we want to remove this assumption, we have to make explicit the  $\beta(\mathbf{r})$  definition given in Eq. (14) and insert it into Eq. (13) thus obtaining:

$$\begin{aligned} \Omega \cdot \nabla \phi_p(\mathbf{r}, E) + \Sigma_t(\mathbf{r}, E) \phi_p(\mathbf{r}, E) = \\ \frac{1}{\bar{k}_p} \sum_m \left[ \chi^{(m)}(E) \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \phi_p(\mathbf{r}, E') \rangle_{E'} - \sum_i \chi_{d,i}^{(m)}(E) \bar{v}_{d,i}^{(m)} \langle \Sigma_f^{(m)}(\mathbf{r}, E') \phi_p(\mathbf{r}, E') \rangle_{E'} \right] \\ + \langle \Sigma_s(\mathbf{r}, E' \rightarrow E) \phi_p(\mathbf{r}, E') \rangle_{E'} \end{aligned} \quad (19)$$

Following the same perturbation approach, i.e. the perturbed system given by Eq. (19) and the unperturbed system given by Eq. (10), we find:

$$\beta_{\text{eff}}^{\text{MC}} = 1 - \frac{\bar{k}_p}{\bar{k}} = \sum_m \sum_i \bar{v}_{d,i}^{(m)} \frac{\langle \langle \phi^+(\mathbf{r}, E) \chi_{d,i}^{(m)}(E) \langle \Sigma_f^{(m)}(\mathbf{r}, E') \phi_p(\mathbf{r}, E') \rangle_{E'} \rangle_E \rangle_{\mathbf{r}}}{\sum_m \langle \langle \phi^+(\mathbf{r}, E) \chi^{(m)}(E) \langle v^{(m)}(E') \Sigma_f^{(m)}(\mathbf{r}, E') \phi_p(\mathbf{r}, E') \rangle_{E'} \rangle_E \rangle_{\mathbf{r}}} \quad (20)$$

Analogously, the classical  $\beta_{\text{eff}}$  definition given in Eq. (7) is an “improved” PT first-order approximation of the relationship  $1 - (\bar{k}_p/\bar{k})$  given in Eq. (20). Also in this case Eqs. (19) and (20) are intimately coupled, in the sense that the coherent prompt flux  $\phi_p$  to be used in the  $\beta_{\text{eff}}$  definition given by Eq. (20) is the eigenfunction of Eq. (19). The previous elaboration can be summarised in the following way:

- As introduced in the previous section, the classical  $\beta_{\text{eff}}$  definitions, like those given in Eqs. (7) and (8), which do not require any prompt flux calculation, are “improved” PT first-order approximations of the relationship  $1 - (\bar{k}_p/\bar{k})$  used by Monte Carlo calculations.
- The relationship  $1 - (\bar{k}_p/\bar{k})$  provides a  $\beta_{\text{eff}}$  estimate coherent with the assumptions made when performing the prompt flux calculation, i.e. Eq. (17) provides the estimate given by Eq. (18), and Eq. (19) provides the estimate given by Eq. (20).

## ERANOS formalism and calculation set-up

The following  $\beta_{\text{eff}}$  formulation is adopted in ERANOS [8] (given in the energy multi-group scheme, with  $g$  as energy group index):

$$\beta_{\text{eff}} = \sum_m \sum_i \bar{v}_{d,i}^{(m)} \frac{\left\langle \sum_g \varphi_g^+(\mathbf{r}) \bar{\chi}_{d,i,g} \sum_{g'} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{g'}(\mathbf{r}) \right\rangle_{\mathbf{r}}}{\left\langle \sum_g \varphi_g^+(\mathbf{r}) \bar{\chi}_g \sum_m \sum_{g'} v_{g'}^{(m)} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{g'}(\mathbf{r}) \right\rangle_{\mathbf{r}}} \quad (21)$$

where  $\bar{\chi}_{d,i,g}$  is a unique delayed neutron spectrum (derived from basic data) for all the fissile isotopes  $m$ , delayed neutron family  $i$ , and:

$$\bar{\chi}_g = \frac{\sum_m \chi_g^{(m)} \sum_{g'} v_{g'}^{(m)} \Sigma_{f,g'}^{(m)} \psi_{g'}}{\sum_m \sum_{g'} v_{g'}^{(m)} \Sigma_{f,g'}^{(m)} \psi_{g'}} \quad (22)$$

The quantity  $\bar{\chi}_g$  is a unique fission spectrum, used by ERANOS for spatial calculations, for all the fissile isotopes.  $\psi_g$  is a weighting flux obtained by the fuel cell cross-section calculations. In Eq. (21) the direct flux and the adjoint flux are solutions of the following eigenvalue equations, respectively:

$$\mathbf{\Omega} \cdot \nabla \varphi_g(\mathbf{r}) + \Sigma_{t,g}(\mathbf{r}) \varphi_g(\mathbf{r}) = \frac{1}{k} \bar{\chi}_g \sum_m \sum_{g'} v_{g'}^{(m)} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{g'}(\mathbf{r}) + \sum_{g'} \Sigma_{s,g' \rightarrow g}(\mathbf{r}) \varphi_{g'}(\mathbf{r}) \quad (23)$$

$$-\mathbf{\Omega} \cdot \nabla \varphi_g^+(\mathbf{r}) + \Sigma_{t,g}(\mathbf{r}) \varphi_g^+(\mathbf{r}) = \frac{1}{k} \sum_m v_g^{(m)} \Sigma_{f,g}^{(m)}(\mathbf{r}) \sum_{g'} \bar{\chi}_{g'} \varphi_{g'}^+(\mathbf{r}) + \sum_{g'} \Sigma_{s,g \rightarrow g'}(\mathbf{r}) \varphi_{g'}^+(\mathbf{r}) \quad (24)$$

It can be seen that, besides the approximation on the delayed neutron spectrum and on the total fission spectrum, Eq. (21) is the energy multi-group version of Eq. (7). Considering as perturbed state with respect to the reference state in Eq. (23) the solution of the following equation:

$$\begin{aligned} \mathbf{\Omega} \cdot \nabla \varphi_{p,g}(\mathbf{r}) + \Sigma_{t,g}(\mathbf{r}) \varphi_{p,g}(\mathbf{r}) &= \frac{1}{k_p} \sum_m \sum_{g'} \left[ \bar{\chi}_g v_{g'}^{(m)} - \sum_i \bar{\chi}_{d,i,g} \bar{v}_{d,i}^{(m)} \right] \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{p,g'}(\mathbf{r}) \\ &+ \sum_{g'} \Sigma_{s,g' \rightarrow g}(\mathbf{r}) \varphi_{p,g'}(\mathbf{r}) \end{aligned} \quad (25)$$

we obtain as exact perturbation:

$$\beta_{\text{eff}}^{\text{MC}} = 1 - \frac{k_p}{k} = \sum_m \sum_i \bar{v}_{d,i}^{(m)} \frac{\left\langle \sum_g \varphi_g^+(\mathbf{r}) \bar{\chi}_{d,i,g} \sum_{g'} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{p,g'}(\mathbf{r}) \right\rangle_{\mathbf{r}}}{\left\langle \sum_g \varphi_g^+(\mathbf{r}) \bar{\chi}_g \sum_m \sum_{g'} v_{g'}^{(m)} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{p,g'}(\mathbf{r}) \right\rangle_{\mathbf{r}}} \quad (26)$$

On the basis of the theory outlined in the previous section, Eq. (21) is the “improved” PT first-order approximation of Eq. (26). Unfortunately ERANOS cannot calculate the solution to Eq. (25), and consequently Eq. (26), because, as mentioned before, it uses a unique fission spectrum for all the fissile isotopes (and in Eq. (25) we have both  $\bar{\chi}_g$  and the various  $\bar{\chi}_{d,i,g}$ ).

In addition, to use (for our scopes) the  $\beta_{\text{eff}}$  modules built-in in ERANOS we need a further assumption, i.e. we have to evaluate:

$$\bar{v}^{(m)} = \frac{\sum_{g'} v_{g'}^{(m)} \Sigma_{f,g'}^{(m)} \Psi_{g'}}{\sum_{g'} \Sigma_{f,g'}^{(m)} \Psi_{g'}} \quad (27)$$

with  $\Psi_g$  as weighting flux obtained by the cell cross-sections calculation. Thus, our new reference state is given by the following equation:

$$\Omega \cdot \nabla \varphi_g(\mathbf{r}) + \Sigma_{t,g}(\mathbf{r}) \varphi_g(\mathbf{r}) = \frac{1}{k} \bar{\chi}_g \sum_m \bar{v}^{(m)} \sum_{g'} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{g'}(\mathbf{r}) + \sum_{g'} \Sigma_{s,g' \rightarrow g}(\mathbf{r}) \varphi_{g'}(\mathbf{r}) \quad (28)$$

with the associated adjoint:

$$-\Omega \cdot \nabla \varphi_g^+(\mathbf{r}) + \Sigma_{t,g}(\mathbf{r}) \varphi_g^+(\mathbf{r}) = \frac{1}{k} \sum_m \bar{v}^{(m)} \Sigma_{f,g}^{(m)}(\mathbf{r}) \sum_{g'} \bar{\chi}_{g'} \varphi_{g'}^+(\mathbf{r}) + \sum_{g'} \Sigma_{s,g \rightarrow g'}(\mathbf{r}) \varphi_{g'}^+(\mathbf{r}) \quad (29)$$

where:<sup>3</sup>

$$\bar{\chi}_g = \frac{\sum_m \chi_g^{(m)} \bar{v}^{(m)} \sum_{g'} \Sigma_{f,g'}^{(m)} \Psi_{g'}}{\sum_m \bar{v}^{(m)} \sum_{g'} \Sigma_{f,g'}^{(m)} \Psi_{g'}} \quad (30)$$

Then setting:

$$\bar{\chi}_{p,g} = \frac{\sum_m \left[ \bar{\chi}_g \bar{v}^{(m)} - \sum_i \bar{\chi}_{d,i,g} \bar{v}_{d,i}^{(m)} \right] \sum_{g'} \Sigma_{f,g'}^{(m)} \Psi_{g'}}{\sum_m \left[ \bar{v}^{(m)} - \sum_i \bar{v}_{d,i}^{(m)} \right] \sum_{g'} \Sigma_{f,g'}^{(m)} \Psi_{g'}} \quad (31)$$

Eq. (25) becomes:

$$\begin{aligned} \Omega \cdot \nabla \varphi_{p,g}(\mathbf{r}) + \Sigma_{t,g}(\mathbf{r}) \varphi_{p,g}(\mathbf{r}) &= \frac{1}{k_p} \bar{\chi}_{p,g} \sum_m \left[ \bar{v}^{(m)} - \sum_i \bar{v}_{d,i}^{(m)} \right] \sum_{g'} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{p,g'}(\mathbf{r}) \\ &+ \sum_{g'} \Sigma_{s,g' \rightarrow g}(\mathbf{r}) \varphi_{p,g'}(\mathbf{r}) \end{aligned} \quad (32)$$

Considering now Eq. (32)<sup>4</sup> as the perturbed state with respect to the reference state in Eq. (28), taking into account Eq. (31), we obtain:

$$\beta_{\text{eff}}^{\text{MC}} = 1 - \frac{k_p}{k} = \sum_m \sum_i \bar{v}_{d,i}^{(m)} \frac{\left\langle \sum_g \varphi_g^+(\mathbf{r}) \bar{\chi}_{d,i,g} \sum_{g'} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{p,g'}(\mathbf{r}) \right\rangle_{\mathbf{r}}}{\left\langle \sum_g \varphi_g^+(\mathbf{r}) \bar{\chi}_g \sum_m \bar{v}^{(m)} \sum_{g'} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{p,g'}(\mathbf{r}) \right\rangle_{\mathbf{r}}} \quad (33)$$

3. Of course, because of Eq. (27), Eq. (30) provides the same result as Eq. (22).

4. On the basis of Eq. (9), Eq. (32) is equivalent to:

$$\Omega \cdot \nabla \varphi_{p,g}(\mathbf{r}) + \Sigma_{t,g}(\mathbf{r}) \varphi_{p,g}(\mathbf{r}) = \frac{1}{k_p} \bar{\chi}_{p,g} \sum_m \left[ 1 - \sum_i \beta_i^{(m)} \right] \bar{v}^{(m)} \sum_{g'} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{p,g'}(\mathbf{r}) + \sum_{g'} \Sigma_{s,g' \rightarrow g}(\mathbf{r}) \varphi_{p,g'}(\mathbf{r})$$

The corresponding “improved” PT first-order approximation of Eq. (33) is:

$$\beta_{\text{eff}} = \sum_m \sum_i \bar{v}_{d,i}^{(m)} \frac{\left\langle \sum_g \varphi_g^+(\mathbf{r}) \bar{\lambda}_{d,i,g} \sum_{g'} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{g'}(\mathbf{r}) \right\rangle_{\mathbf{r}}}{\left\langle \sum_g \varphi_g^+(\mathbf{r}) \bar{\lambda}_g \sum_m \bar{v}^{(m)} \sum_{g'} \Sigma_{f,g'}^{(m)}(\mathbf{r}) \varphi_{g'}(\mathbf{r}) \right\rangle_{\mathbf{r}}} \quad (34)$$

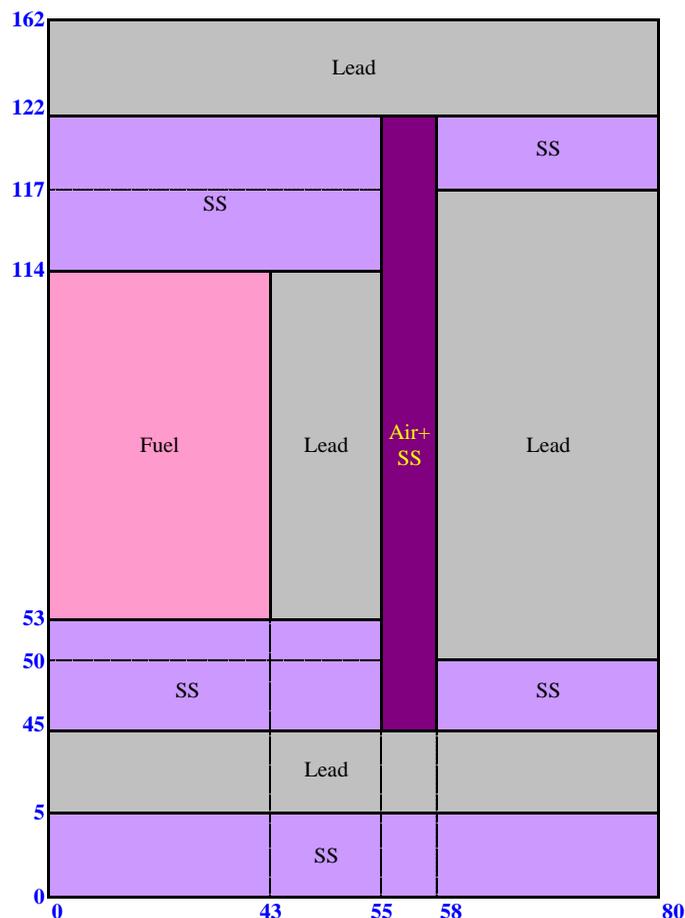
Eq. (32), both sides of Eqs. (33) and (34) can be calculated by ERANOS.

### ERANOS results

The GUINEVERE system, shown in Figure 1, is selected to perform the numerical analysis. The fuel fissile zone is made of uranium 30% (weight) enriched in <sup>235</sup>U. The fuel matrix has the following volumetric fractions: uranium 17%, stainless steel 16%, lead 60% and air 7%. The zone air+ (core lattice) sheet has the following volumetric fractions: stainless steel 93% and air 7%. The RZ system is a simplified schematisation of the GUINEVERE start-up (at critical) configuration (after start-up the central zone is replaced by the beam tube of the deuteron accelerator delivering, in continuous or pulsed mode, 14 MeV neutrons by deuterium-tritium reactions).

**Figure 1: Simplified RZ model of the GUINEVERE start-up (at critical) configuration**

Dimensions, not in scale, are given in cm



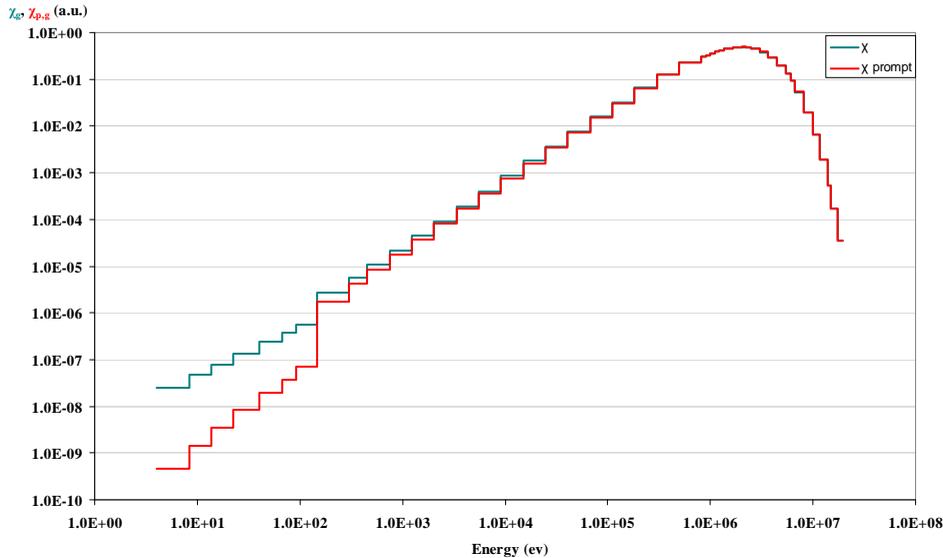
ERANOS transport calculations are performed with 49 energy groups (see Appendix),  $P_0$  transport approximation, angular quadrature  $S_4$  (module BISTRO [6]). The neutron data library JEFF-3.1 [7] is used for the calculations. In Table 1 the delayed neutron data  $\bar{\nu}_{d,i}^{(m)}$  [9] used for the analysis are shown. Delayed neutron spectra  $\bar{\chi}_{d,i,g}$  are the ones built-in in ERANOS and taken from [9].

**Table 1: Delayed neutron data  $\bar{\nu}_{d,i}^{(m)}$  [9]**

Family	1	2	3	4	5	6	7	8	Sum
$^{235}\text{U}$	0.00055	0.00245	0.00162	0.00326	0.00509	0.00152	0.00142	0.00039	0.01630
$^{238}\text{U}$	0.00039	0.00484	0.00174	0.00637	0.01367	0.00921	0.00595	0.00433	0.04650

The following  $\bar{\nu}^{(m)}$  values were obtained by Eq. (27):  $\bar{\nu}^{(235)} = 2.49$  and  $\bar{\nu}^{(238)} = 2.75$ .<sup>5</sup> In Figure 2 a comparison between the total fission spectrum  $\bar{\chi}_g$  obtained by Eq. (30) and the prompt fission spectrum  $\bar{\chi}_{p,g}$  obtained by Eq. (31) is shown. A synthesis of the results is reported in Table 2.

**Figure 2: Total fission spectrum  $\bar{\chi}_g$  [Eq. (30)] and prompt fission spectrum  $\bar{\chi}_{p,g}$  [Eq. (31)]**



**Table 2: Synthesis of the results**

Case	Equation	k	$k_p$	$\beta_{\text{eff}}$ (pcm)	$1 - (k_p/k)$ (pcm)
(a)	(21) for $\beta_{\text{eff}}$			723	
(b)	(23) for $\varphi$	1.01955			
(c)	(24) for $\varphi^+$	1.01955			
(d)	(28) for $\varphi$	1.02149			
(e)	(29) for $\varphi^+$	1.02149			
(f)	(32) for $\varphi_p$		1.01402		
(g)	(33) for $1 - (k_p/k)$				732
(h)	(33) for $\beta_{\text{eff}}$ (using $\varphi_p$ )			732	
(i)	(34) for $\beta_{\text{eff}}$ (using $\varphi$ )			731	

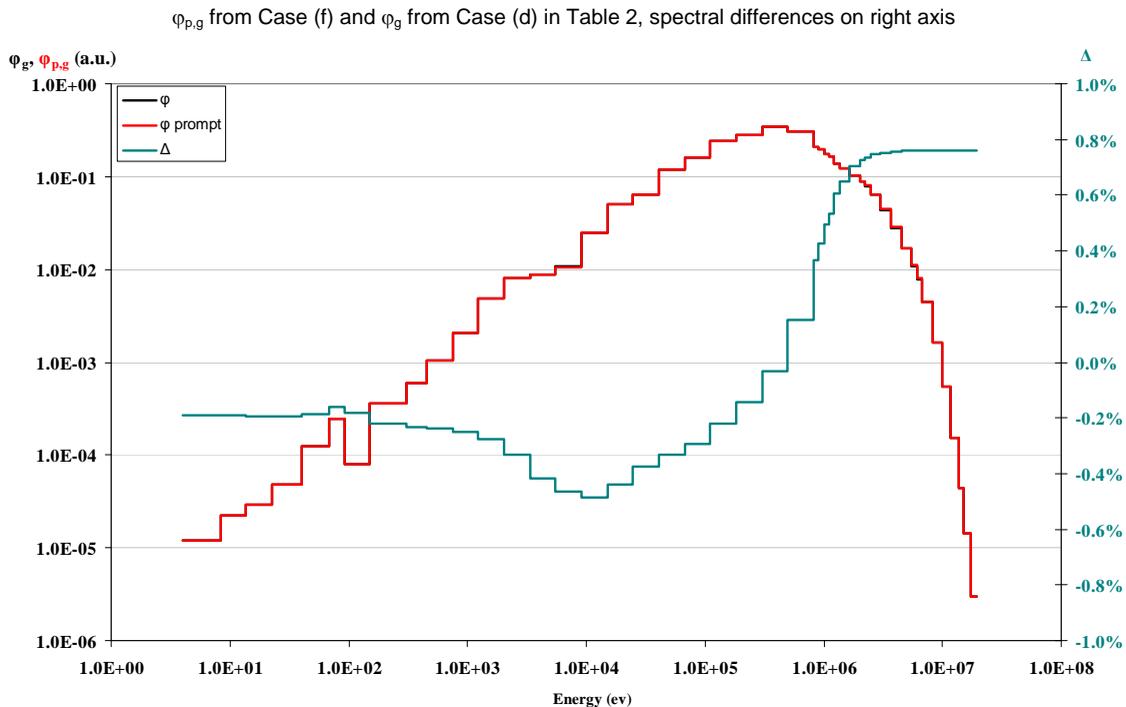
5. This high  $\bar{\nu}^{(238)}$  value is due to the fact that about 61% of  $^{238}\text{U}$  total fissions occur in the energy range 2–8 MeV, where  $\bar{\nu}^{(238)}$  values span from 2.67 to 3.48 (only about 5% of  $^{235}\text{U}$  total fissions occur above 2 MeV).

Case (a) is the classical  $\beta_{\text{eff}}$  formulation adopted in ERANOS, with  $\varphi$  and  $\varphi^+$  from Cases (b) and (c) respectively. Cases (d) and (e) differ from (b) and (c) for the presence of  $\bar{\nu}^{(m)}$  in fission production terms, and this approximation provides a difference of +186 pcm with respect to the standard treatment of the fission production terms. The prompt Case (f), again with the presence of  $\bar{\nu}^{(m)}$  in the fission production terms, provides a difference of -721 pcm ( $-\beta_{\text{eff}}$ ) with respect to Cases (d) and (e).

Case (g), representing the approach used by Monte Carlo calculations, provides exactly the value by Case (h), i.e. the last term in Eq. (33). This numerical result confirms the theoretical analysis described above, linking the Monte Carlo formulation to the PT approach. Finally, Case (i) shows that the replacement of the prompt flux in Case (h) by the total flux does not affect appreciably the  $\beta_{\text{eff}}$  value (at least for the fast system considered).

Actually, the differences between  $\varphi$  and  $\varphi_p$ ,<sup>6</sup> from Cases (d) and (f) in Table 2, respectively, are not remarkable, as can be seen from Figure 3 (where  $\varphi$  and  $\varphi_p$  are not distinguishable).

**Figure 3: Total and prompt fluxes, together with spectral differences  $\Delta = (\varphi_{p,g} - \varphi_g)/\varphi_g$**



## Conclusions

When evaluating  $\beta_{\text{eff}}$  by the relationship  $1 - (k_p/k)$ , the quality of the obtained result depends on the quality of the description of the delayed neutron emission assumed in the  $k_p$  prompt calculation. It could seem evident, but if we look at the standard route used in ERANOS (and in general in deterministic codes) to calculate  $\beta_{\text{eff}}$ , by a formulation which does not require any prompt flux calculation, we realise that great detail is given to the characteristics of the delayed neutron emission through the  $\beta_{\text{eff}}$  formulation, and without the need to transpose such a detail into a full system prompt neutron calculation. For example, in the ERANOS case it is not possible to set up a prompt calculation having the same quality of the delayed neutron emission

6.  $\varphi$  and  $\varphi_p$  in Figure 3 are spatial average values over the core volume. Same area under  $\varphi$  and  $\varphi_p$  curves.

description available in the  $\beta_{\text{eff}}$  formulation. And in fact, in our case, we were obliged to lower the information about the delayed neutron emission in the  $\beta_{\text{eff}}$  definition in order to run the corresponding prompt calculation.

Following the perturbation theory approach, a rigorous relationship may be established between the  $\beta_{\text{eff}}$  evaluation by the formula  $1 - (k_p/k)$ , used in Monte Carlo codes, and the corresponding  $\beta_{\text{eff}}$  calculation by the classical formulation involving direct and adjoint fluxes, and it has been shown how the classical formulation can be considered an “improved” PT first-order formulation of the formula  $1 - (k_p/k)$ . By means of a refined 49 energy group structure, transport computational analysis carried out in a coherent and consistent way, i.e. using the same deterministic code ERANOS and neutron data library JEFF-3.1 for the  $\beta_{\text{eff}}$  evaluation in different ways, the theoretical analysis is numerically confirmed.

Both theoretical and numerical results confirm the effectiveness of the  $\beta_{\text{eff}}$  evaluation by the relationship  $1 - (k_p/k)$ , at least in cases where spectral differences between total and prompt fluxes are negligible with respect to the value of the functionals entering the classical  $\beta_{\text{eff}}$  formulation. For other material configurations more investigations are needed to obtain a detailed quantification of the effects involved.

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### Appendix: 49 energy group scheme

Upper energy (eV)	Group #						
1.96E+07	1	2.470E+06	14	1.11E+05	27	1.49E+02	40
1.73E+07	2	2.23E+06	15	6.74E+04	28	9.17E+01	41
1.49E+07	3	2.02E+06	16	4.09E+04	29	6.79E+01	42
1.38E+07	4	1.65E+06	17	2.48E+04	30	4.02E+01	43
1.16E+07	5	1.35E+06	18	1.50E+04	31	2.26E+01	44
1.00E+07	6	1.22E+06	19	9.12E+03	32	1.37E+01	45
8.19E+06	7	1.11E+06	20	5.53E+03	33	8.32E+00	46
6.70E+06	8	1.00E+06	21	3.35E+03	34	4.000E+00	47
6.07E+06	9	9.07E+05	22	2.03E+03	35	5.40E-01	48
5.49E+06	10	8.21E+05	23	1.23E+03	36	1.00E-01	49
4.49E+06	11	4.98E+05	24	7.49E+02	37		
3.68E+06	12	3.02E+05	25	4.54E+02	38		
3.04E+06	13	1.83E+05	26	3.04E+02	39		

## Experimental validation of reactivity monitoring techniques for power ADS systems to incinerate radioactive wastes\*

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### Abstract

The YALINA-Booster programme was designed within the framework of the IP-EUROTRANS for validation of the reactivity monitoring techniques on ADS systems. The main objective was the development of a validated methodology to be used in future power ADS. For this purpose, YALINA-Booster is a zero-power subcritical assembly with a fast-thermal neutron spectrum coupled to a D-T neutron generator. The high intensity of the accelerator and the possibility to work in continuous or pulsed mode allowed performing standard pulsed neutron experiments, exploring the current-to-flux relationship and performing beam trip experiments. In addition, it has provided the opportunity to test the electronic chains in current mode, corresponding to the most probable operation mode in a power ADS.

Pulsed neutron source (PNS) experiments, previously validated in experiments as MUSE-4, have been carried out to achieve the reference reactivity values for each configuration studied. The techniques applied were the Sjöstrand area method and the prompt neutron decay slope fitting technique. The results obtained from both methods were affected by spatial dependences due to the assembly heterogeneity. These spatial dependences have been corrected using a novel technique based on Monte Carlo simulations.

For the first time, the absolute reactivity values of a subcritical system were determined by imposing short millisecond-scale interruptions to the continuous deuterium beam current (beam trips). This technique provided the possibility to monitor the reactivity values on each second using the source jerk methodology.

Also for the first time, the fast evolution of the neutron flux intensity within the subcritical assembly was measured using fission chambers operating in current mode. This required the development of specific electronic chains specifically designed for this experimental campaign. In order to test the validity of the experimental results, these reactivity values were demonstrated to be compatible with those obtained by standard PNS techniques.

Finally, on-line monitoring of the reactivity has been achieved using the current-to-flux technique. This method relies on the continuous monitoring of the deuteron accelerator current, the neutron source intensity and the neutron flux within the reactor, which is not guaranteed to be proportional to the beam current. With this methodology, relative changes in the subcritical assembly behaviour were possible to be identified in time intervals as short as one millisecond.

\* The full paper being unavailable at the time of publication, only the abstract is included.



## Reaction cross-sections, fission yields and prompt neutron emission from actinide targets

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### Abstract

*A thorough understanding of the fission process for actinides is becoming an important necessity for the modelling of innovative concepts for radioactive waste minimisation based on either accelerator-driven systems or fast reactors, e.g. Generation IV systems. There is a need for improved nuclear data especially in the fast neutron energy range and for minor actinides, which have not been investigated in great detail so far.*

*Investigations for the understanding of the fission process are one focus of the research activities at JRC-IRMM. Recently, fission fragment properties (mass, kinetic energy and angular distributions) for the isotopes  $^{238}\text{U}$  and  $^{234}\text{U}$  have been studied to improve the knowledge of fission yield data in the threshold region of the neutron-induced fission cross-section. These new and precise data have been used to improve neutron-induced reaction modelling for  $^{238}\text{U}(n,f)$ .*

*The prompt neutron emission in nuclear fission has also been studied for the spontaneous fission of  $^{252}\text{Cf}$ , showing for the first time no drop in neutron multiplicity for low total kinetic energies as expected by theory. Also, the prompt fission neutron spectrum of  $^{235}\text{U}$  is being investigated in detail, both at thermal and for higher-energy incoming neutrons. In the case of  $^{235}\text{U}$  the available experimental data are inconsistent with each other and contradictory to theoretical predictions. The new accurate spectral data are used to determine a realistic prompt fission neutron spectrum which is used to calculate critical assemblies and  $K_{\text{eff}}$ .*

*A second part of the work programme at JRC-IRMM focuses on the precise measurement of resonance parameters present in the neutron-induced reaction cross-sections at incident neutron energies up to several hundreds of electronvolts. The present evaluated data for neutron-induced reactions in  $^{241}\text{Am}$  do not fulfil the requirements, as spelled out by the OECD/NEA Subgroup 26 to accurately predict the behaviour of a reactor core while burning minor actinides. Therefore, high resolution capture and transmission measurements were carried out at the time-of-flight facility GELINA on  $^{241}\text{Am}$  in an energy region from thermal energy up to 500 eV. A preliminary analysis indicates an underestimation of the capture cross-section in the present evaluations.*

## Introduction

The fission process plays an important role in many applications in nuclear science as well as in basic nuclear physics. Despite the fact that fission has been known of for more than 70 years the present description of the fission process remains largely dominated by phenomenology. Hence, consistent models to describe the fission process, i.e. the properties of the fissioning system, fission dynamics and fragment distributions, are still lacking. To this end it is important to investigate the fission process in depth from the experimental perspective to provide the necessary input parameters for the improvement of nuclear fission models.

A thorough understanding of the fission process for actinides is becoming an important necessity for the modelling of innovative concepts for radioactive waste minimisation based on either accelerator-driven systems or fast reactors, e.g. Generation IV systems. There is a need for improved nuclear data especially in the fast neutron energy range and for minor actinides, which have not yet been investigated in great detail.

Investigations for the understanding of the fission process are one focus of the research activities at JRC-IRMM. Examples are presented in the following sections.

A second part of the work programme at JRC-IRMM focuses on the precise measurement of resonance parameters present in the neutron-induced reaction cross-sections at incident neutron energies up to several hundreds of electronvolts. For all reactor cores that contain plutonium,  $^{241}\text{Am}$  is present. This is caused by the radioactive decay of  $^{241}\text{Pu}$ , which will be present as a result of neutron-induced reactions and decays occurring in the core. Therefore precise and accurate cross-section values for the total, capture and fission cross-sections of Am are required for monitoring the performance of the device and for nuclear forensics.

## Experimental results

The JRC-IRMM has a long-standing tradition in the field of neutron-induced fission physics studies and the measurement of resonance parameters. It is especially well-equipped, with world-class facilities such as the high-resolution neutron time-of-flight spectrometer GELINA and the 7 MV Van de Graaff accelerator for quasi-monoenergetic neutron production (MONNET).

Over the recent past several neutron-induced fission reactions have been studied in the energy range up to 6 MeV and for spontaneous fission. Angular, mass and total kinetic energy (TKE) distributions have been investigated for  $^{234,238}\text{U}(n,f)$ , prompt neutron emission in spontaneous fission of  $^{252}\text{Cf}$  as well as the prompt fission neutron spectrum of  $^{235}\text{U}(n,f)$ . In the following sections the individual isotopes will be discussed and, due to space limitations, only the most important results will be presented.

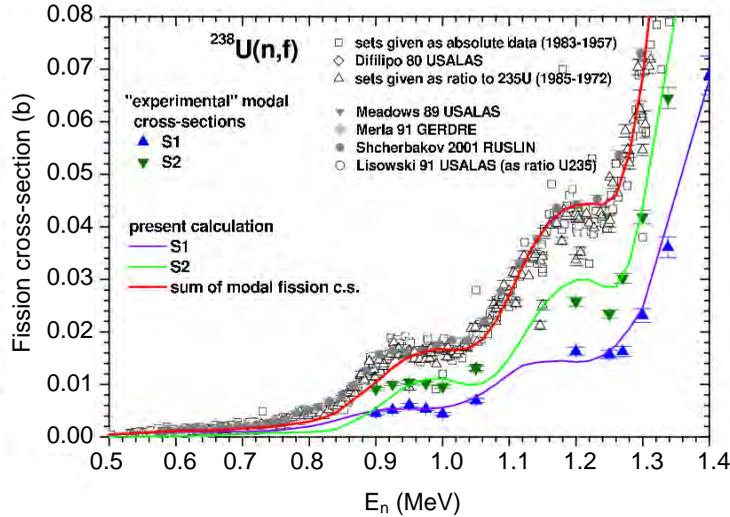
### ***Fission yields for $^{234,238}\text{U}(n,f)$***

The fission fragment properties like mass, angular distribution and TKE have been studied applying the so-called 2E method in the threshold region of the fission cross-section. The goal was to verify the existence of large fluctuations in the fission fragment properties as reported in literature (see references in Ref. [1]). As fission fragment detector a twin Frisch-grid ionisation chamber was used. The neutron beam was supplied by the JRC-IRMM Van de Graaff accelerator using the  $^3\text{H}(p,n)$  and  $^7\text{Li}(p,n)$  reaction. More details about the experiment and the results are given in Ref. [1].

Here we will concentrate on the improved calculation of the reaction cross-sections and branching ratios as a function of the incident neutron energy based on fission fragment data from Ref. [1]. This needs to be compared to the results given in Ref. [2]. Figure 1 shows the improved calculation [3] with improved barrier parameters compared to Ref. [2], based on the parabolic barrier shape approximation. The up and down triangles represent the experimentally

**Figure 1: S1 and S2 modal cross-section as a function of incident neutron energy in the vicinity of the vibrational sub-barrier resonances**

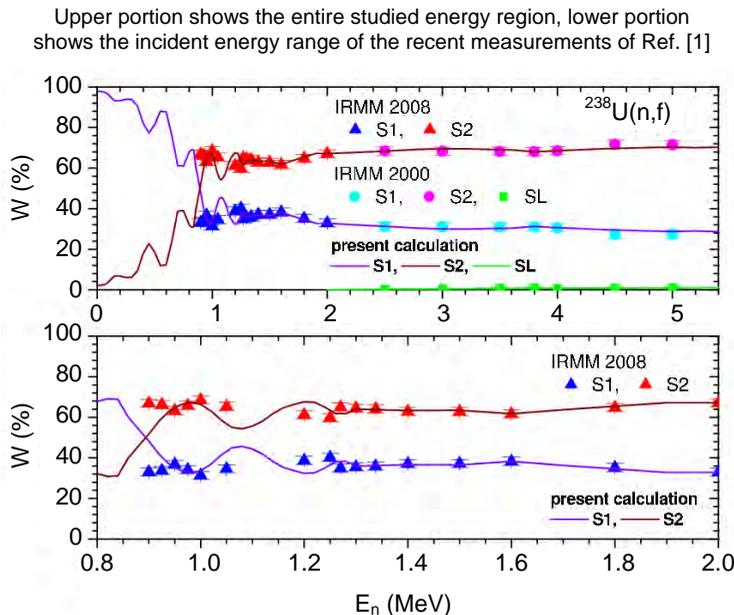
The results of the calculations as well as their sum are depicted in comparison with experimental data



deduced modal fission cross-sections based on modal weights of Ref. [1], the different full lines the calculated modal fission cross-section and their sum. The open symbols represent experimental fission cross-section data from the EXFOR data library.

Figure 2 represents the modal weights of the so-called Standard I and II modes as a function of incident energy. The lines are the predictions of the model [3] compared to the experimental findings (full triangles). Compared to the former calculation (for details see Ref. [2]) the improved version shows less fluctuation of the modal weights around the vibrational resonances in the threshold of the fission cross-section.

**Figure 2: Calculated modal weights for the standard modes in  $^{239}\text{U}$  (full lines) in comparison with experimental data (full symbols)**

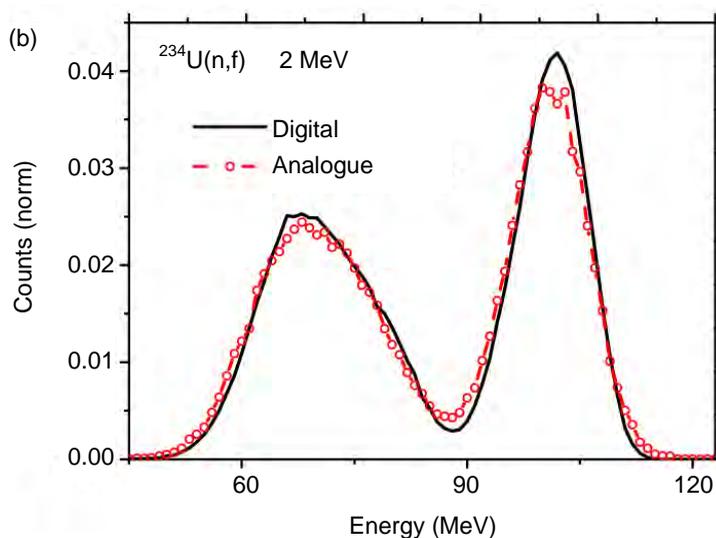


The investigation of the reaction  $^{234}\text{U}(n,f)$  has also been done to study in these isotopes the influence of the strong vibrational resonance in the threshold of the fission cross-sections on the fission fragment properties. Further experience was gained in the use of the digital signal processing technique applied to fission fragment spectroscopy.

Analysis of the experimental data is still in progress, but initial results are available. The measurement was done both in the traditional analogue technique using conventional analogue electronic modules and by digitising the ionisation chamber signals using fast waveform digitisers. A comparison of the results and the benefits of the digital technique has already been published [4]. Figure 3 shows as an example the improvement in the energy distribution of the fragments comparing the results obtained using the digital signal processing routines (full line) with the traditional analogue technique (symbols connected by line).

**Figure 3: Energy distribution for  $^{234}\text{U}(n,f)$  at 2 MeV neutron energy, for both analogue and digital data acquisition**

Due to the event-by-event pile-up correction in the digital case the energy resolution was significantly improved



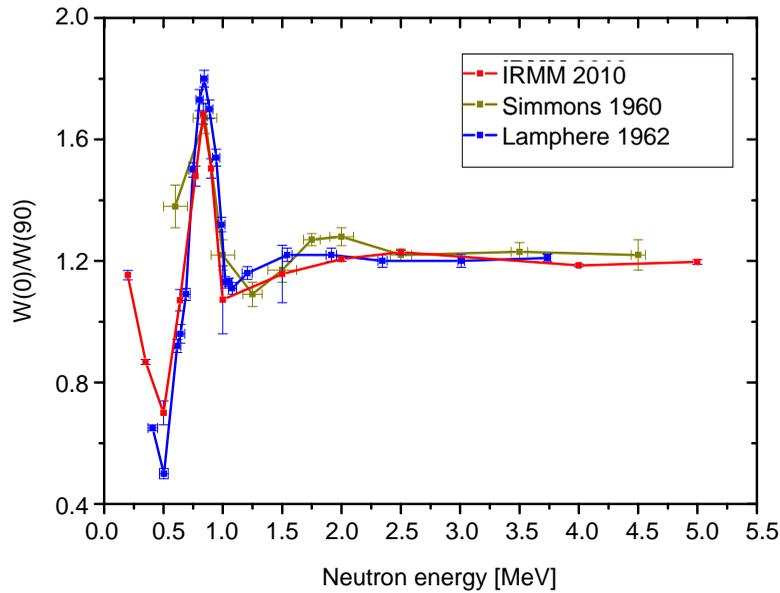
Another preliminary result is shown in Figure 4, that of the very strong angular anisotropy observed for this nucleus in the vicinity of the vibrational resonance in the threshold of the fission cross-section in comparison with literature data. We have confirmed the strong drop in anisotropy below 800 keV of Lamphere et al. [5]. Further analysis is in progress.

### **Prompt neutron emission in fission of $^{252}\text{Cf}(SF)$**

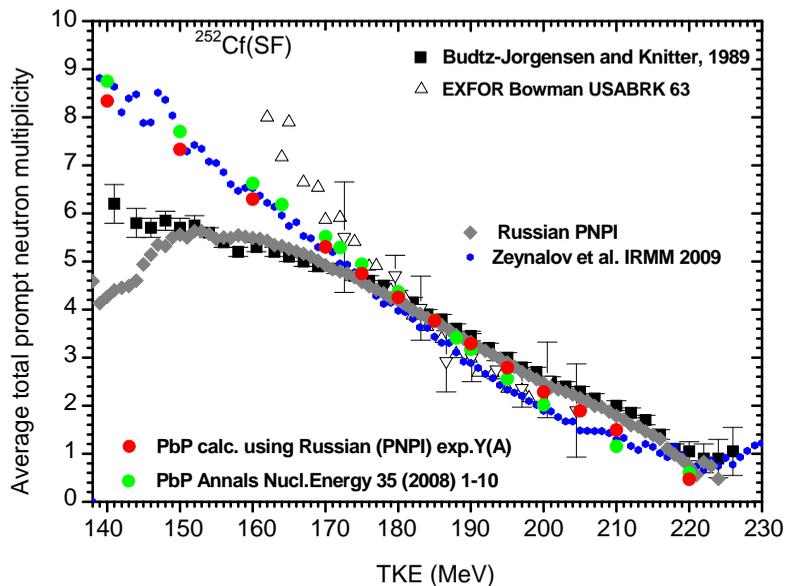
We have repeated the pioneering experiment of Budtz-Jorgensen and Knitter [6] to verify the results with improved statistics and using for the first time a digital data acquisition system for both the fission fragment and prompt neutron emission part. The data analysis is still in progress but first results show the complete absence of a reduction of the prompt neutron multiplicity at low total kinetic energies (TKE) and a different slope of this function towards high TKE compared to Ref. [6] (see Figure 5).

Also recent calculations within the point by point (PbP) model are shown in comparison giving the same results as the present experimental data. In both cases the new results do not confirm the older results in terms of the slope of the distribution and reduction in neutron multiplicity below, say, a TKE of 160 MeV.

**Figure 4: Anisotropy of the fission fragment angular distribution for  $^{234}\text{U}(n,f)$  as a function of incident neutron energy**



**Figure 5: Neutron multiplicity as a function of total kinetic energy (TKE) of the fission fragments for  $^{252}\text{Cf}(SF)$  in comparison with literature data**



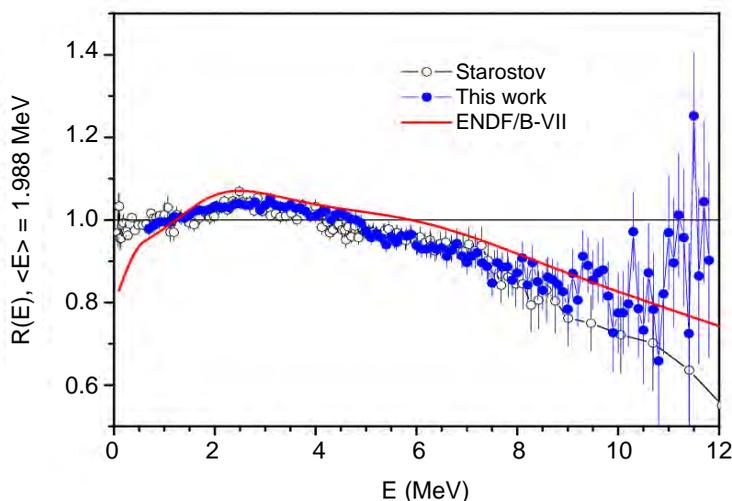
### Prompt fission neutron spectrum of $^{235}\text{U}(n, f)$

The prompt fission neutron spectrum (PFNS) has been measured at the cold-neutron beam of the Budapest Research Reactor [7]. The experiment was supported within the FP6 European Facilities for Nuclear Data Measurements (EFNUDAT) project.

Three NE213-equivalent neutron scintillation detectors surrounded by large lead and lithium-carbonated paraffin shieldings have been used simultaneously together with a double Frisch-grid ionisation chamber for the fission fragment detection. The detailed characterisation of the detectors and a description of the experiment is given in Refs. [7, 9]. Here, only the resulting PFNS is presented in Figure 6 relative to a Maxwellian distribution with mean energy  $E = 1.988$  MeV. It is obvious that the measured spectrum (symbols) does not agree with the calculation within the so-called Los Alamos model (full line) [10]. A perfect agreement over the full energy range covered in the present study is observed compared to the literature values of Starostov *et al.* [8] (see Figure 6).

**Figure 6: Ratio of the PFNS for  $^{235}\text{U}(n,f)$  to a Maxwellian distribution with  $T = 1.988$  MeV**

The new experimental points (full symbols) are compared to Ref. [8] and to the evaluation based on the Madland-Nix model [10]



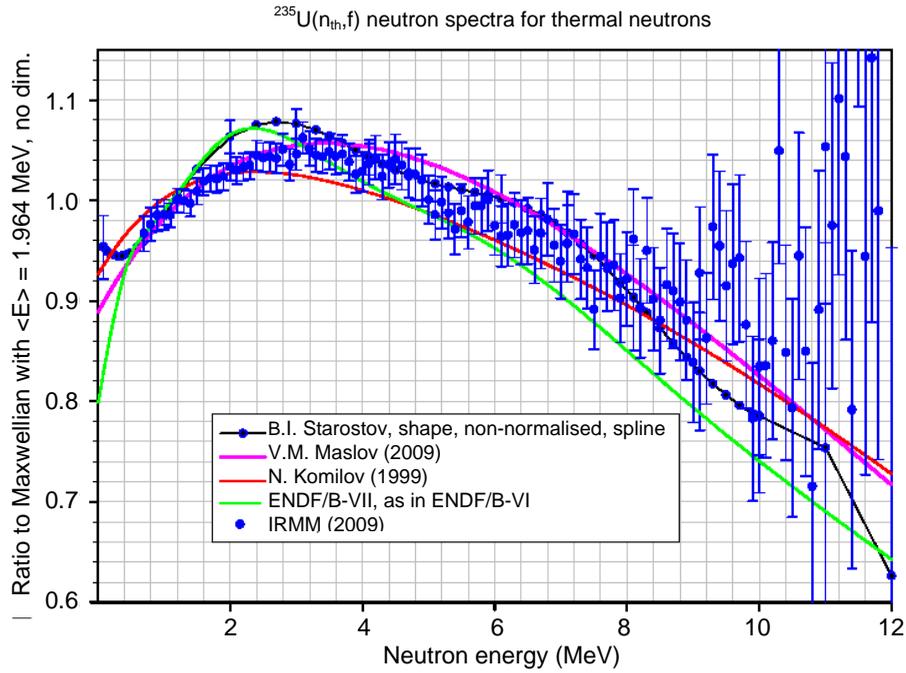
In view of the fact that the influence of the  $^{235}\text{U}(n,f)$  PFNS and its imposed uncertainties on the thermal and fast reactor neutronics is quite appreciable as estimated in Ref. [11], a new spectrum calculation is made by V. Maslov [12] using our new experimental data (see Figure 7). Using this new spectrum V. Maslov *et al.* [13] did an analysis of different critical assemblies and calculated the respective  $K_{\text{eff}}$  values. A selection of intermediate-enriched uranium assemblies is given in Figure 8, which demonstrates the sensitivity to different PFNS representations. For the Big Ten core the criticality obtained using ENDF/B-VII was attained by tuning the neutron capture cross-section of  $^{238}\text{U}$  in the hundreds of keV range. With a realistic PFNS as used by V. Maslov, the response is quite different. The obvious discrepancy between the PFNS model and experimental results triggered an IAEA co-ordinated research project on PFNS for actinides to improve the understanding of this important physical quantity.

## Resonance parameters

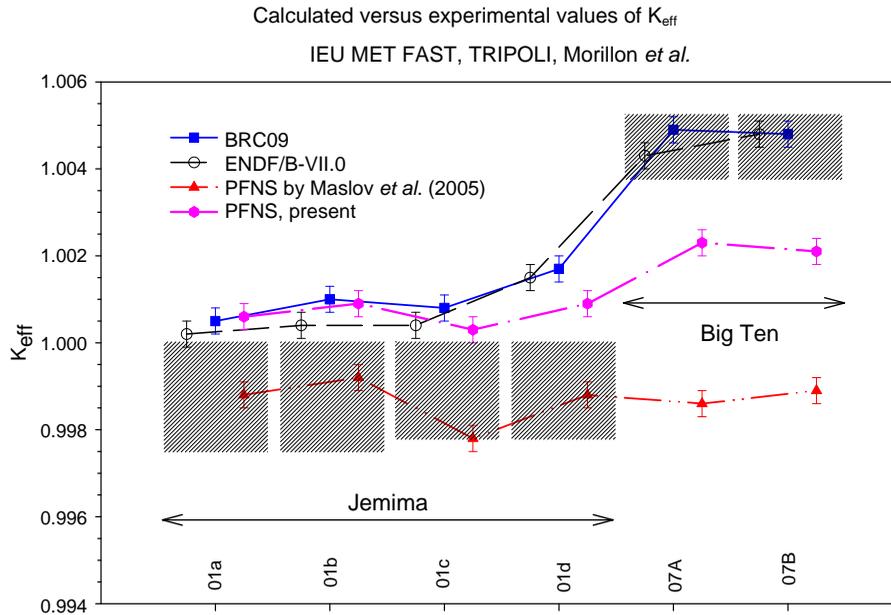
During the last decade four new measurements [14-17] have been devoted to determine the thermal capture cross-section of  $^{241}\text{Am}$ . The values of these four measurements are in agreement with each other, but basically disagree with the evaluated data files, except for JEFF-3.1. The latest measurement by Jandel *et al.* [14] reported resonance parameters with an average total width smaller by a few per cent. The reported  $\Gamma_n$  were also slightly higher compared to previously reported values. The ratio between the neutron widths reported by Jandel *et al.* [14] and some previous experiments, as depicted in Figure 9, shows an energy dependence. Possible explanations could be sample inhomogeneities or neutron flux effects. Therefore another set of independent experiments would be desirable for a reliable new evaluation.

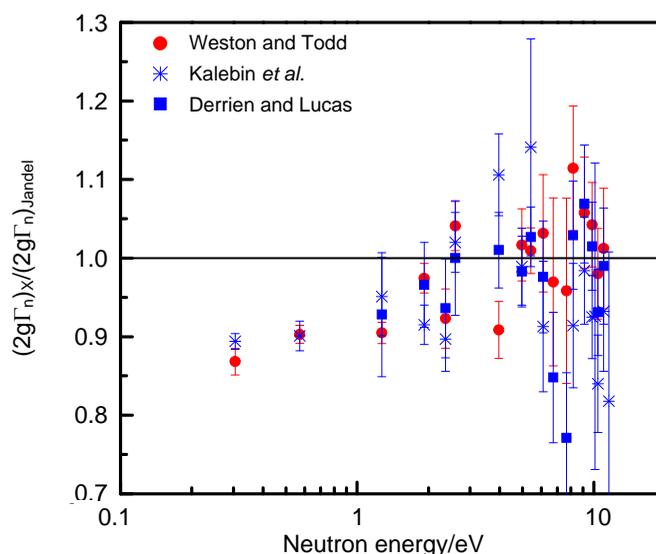
**Figure 7: Ratio of the PFNS for  $^{235}\text{U}(n,f)$  to a Maxwellian distribution with  $T = 1.964$  MeV**

The new experimental points (full symbols) are compared to Ref. [8] and to the evaluation based on the Madland-Nix model [10]; a new calculation of the spectrum by V. Maslov is also given



**Figure 8: Prompt fission neutron spectrum sensitivity for IEU-MET-FAST benchmark**



**Figure 9: Ratio of  $\Gamma_n$  from previous experiments to that of Jandel *et al.* [14]**

The experiments were performed at GELINA, the pulsed neutron source JRC-IRMM. GELINA is a linear accelerator that accelerates electrons up to 150 MeV and has a peak current of 12 A. The use of a post-acceleration magnet allows for electron pulses as short as 1 ns. The repetition rate of these pulses can be varied between 10-800 Hz. The electron beam hits a mercury-cooled uranium target and bremsstrahlung is produced. Consequently neutrons with energies of a few MeV are created by  $(\gamma, xn)$ -reactions and photofission. Two beryllium containers filled with water and mounted above and below the neutron production target are used to moderate the neutrons.

The software package AGS [18] was used to derive the transmission and the capture yields from the registered data. This code performs the most important corrections, such as dead-time correction, background subtraction, etc., with a full propagation of the covariance matrix, starting with the statistical uncertainties of the counting statistics.

Sample homogeneity being of utmost importance for these experiments, a sol-gel technique was used for sample preparation. In approximately 3 g of  $Y_2O_3$ , which was used as the sol-gel matrix, approximately 320 mg of Am was infused. Compared to the commonly used oxide powder samples, a much higher degree of homogeneity will be achieved with such a sample type.

The transmission measurements were performed at flight path 2 of GELINA, a flight path that views the moderator at an angle of  $9^\circ$  with its normal. The chosen flight path length was 26.45 m. The beam diameter was limited to approximately 2 cm, using a combination of lithium-carbonate, copper and nickel collimators. The sample was positioned at a distance of approximately 10 m from the neutron source, so halfway between the neutron target and the detector system. Filters to reduce the intensity of the gamma-flash as well as black resonance filters for estimating the background could be inserted close to the sample position. For neutron detection a 1.27 cm thick lithium-glass scintillator was employed, with a diameter of approximately 10 cm viewed by two photomultiplier tubes.

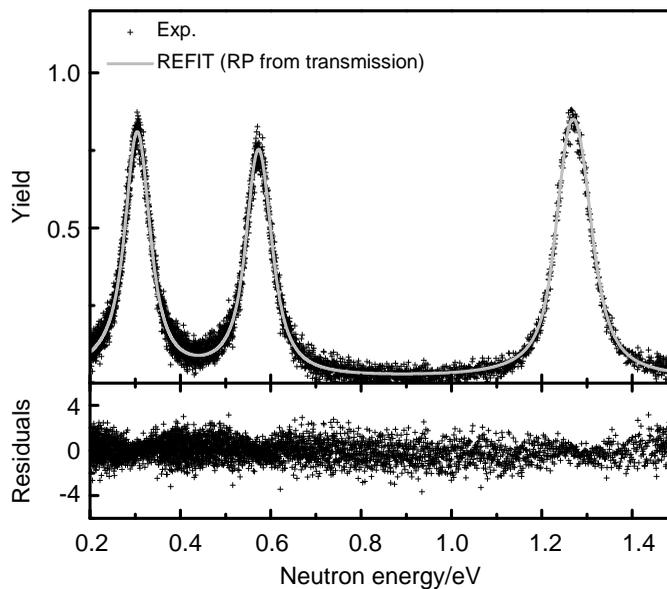
The experiments for determining the capture yield of americium were performed at a flight path length of 10 m with the beam diameter at the sample station reduced to approximately 75 mm using lithium-carbonate plus resin, copper and lead collimators. The detection of the photons that were produced in the capture event is accomplished by two cylindrical  $C_6D_6$  detectors (NE230). These detectors are mounted at an angle of  $125^\circ$  to reduce the effects caused by the anisotropy of the dipole radiation. The pulse height weighing technique was used to create a detector response proportional to the energy of the registered gamma. A description of the method and of the calculation of the weighing function can be found in Ref. [19].

In both set-ups, the stations were air conditioned to keep the samples at a constant temperature and to minimise a drift of the electronics.

An accurate determination of the neutron energies requires a careful measurement of the flight path length. Therefore the flight path length for transmission was calibrated using the energy of 6.674 eV resonance of  $^{238}\text{U}$  as given in the ENDF/B-VII evaluated data file. This gave an effective flight path length of  $(26.444 \pm 0.006)$  m. The uncertainty on the uranium resonance energy was the main contributor to the flight path uncertainty. The neutron energy dependence of the resolution function was determined in a Monte Carlo simulation of the spectrometer set-up [20].

First results with the resonance shape analysis code REFIT [21] indicate that for extracting resonance parameters the crystal lattice model (CLM) is preferred over the free gas model. Comparing the total observed widths, the Doppler broadening and the intrinsic widths, one can conclude that a resonance shape analysis – and therefore a determination of  $E_{\text{res}}$ ,  $\Gamma_n$  and  $\Gamma_\gamma$  is only feasible for low energies. Resonance parameters from the transmission measurement were used unchanged for the calculation of the capture yield (see Figure 10). The good agreement between experimental data and calculation suggests that transmission and capture measurements are consistent with each other. A reliable extraction of resonance parameters should therefore be possible.

**Figure 10: Experimental capture yield compared to R-matrix calculation**



## Conclusions

For  $^{238}\text{U}(n,f)$  both the theoretically predicted as well as the experimentally observed fluctuations in literature in the vibrational resonances in the fission cross-section threshold could not be verified. The input parameters for the statistical model have been revised and adapted to the new results for  $^{238}\text{U}(n,f)$ . For  $^{234}\text{U}(n,f)$  digital data acquisition has resulted in improved fission fragment signals and the strong angular anisotropy of fission fragment emission around the vibrational resonance in the fission cross-section could be verified.

In the spontaneous fission of  $^{252}\text{Cf}$  no reduction of the prompt neutron multiplicity as a function of TKE is observed for the first time, which is in agreement with the conservation law of energy.

For the PFNS of  $^{235}\text{U}(n,f)$  new experimental results have shown that the present theoretical description of the PFNS in terms of the so-called Los Alamos model [10] is not able to reproduce the experimental data. A more realistic PFNS calculation shows deficiencies in other quantities included in  $K_{\text{eff}}$  calculations.

The energies of the first three resonances are in good agreement with the values reported by Jandel *et al.* [14]. The extracted total widths in our experiment seem to be slightly smaller than the values by Jandel *et al.* [14]. Before final values can be reported a detailed characterisation of the sample concerning the impurities, total Am content and possible sample inhomogeneities, remains to be performed.

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## Investigation of nuclear data accuracy for the accelerator-driven system with minor actinide fuel

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### Abstract

Accuracy of nuclear data, especially for minor actinides (MA), is important in neutronics design of advanced reactors for MA transmutation such as an accelerator-driven system (ADS). A benchmark activity to understand current accuracy of neutronic calculations for the ADS was performed in the Co-ordinated Research Project (CRP) on “Analytical and Experimental Benchmark Analyses of Accelerator-driven Systems” held by the International Atomic Energy Agency (IAEA). A commercial-grade ADS with a thermal output of 800 MW was employed in the benchmark and depletion analysis was performed by participants using deterministic or Monte Carlo codes. Results revealed that a discrepancy among  $k$ -effectives by different nuclear data libraries is as large as 2-3%dk even for an initial criticality before burn-up.

Further investigation of the uncertainty was performed using the covariance data in JENDL-3.3 and JENDL-4.0. However, the uncertainty of criticality estimated from JENDL-4.0 was only 1 090 pcm (1.1%dk), which is much smaller than the previous result of the IAEA-CRP. The comparison of covariance data and cross-section differences among the libraries is necessary. The dominant reactions for uncertainty of criticality, void and Doppler reactivity were identified. The capture cross-sections of <sup>237</sup>Np, <sup>241</sup>Am and <sup>243</sup>Am were included in the dominant reactions.

In order to reduce the uncertainty due to such MA, the Transmutation Physics Experimental Facility (TEF-P) with a significant amount of MA is being proposed in the J-PARC project. An expected reduction of the uncertainty by the TEF-P was numerically evaluated using a cross-section adjustment procedure with covariance data from JENDL-3.3. The original uncertainty for  $k$ -effective was about 1.3%. The uncertainty would be reduced from 1.3% to 0.6% if experimental results of the TEF-P and other past critical experiments like the ZPPR were utilised to adjust nuclear data. Therefore, the MA-loaded critical experiment at the TEF-P is important to improve the uncertainty for MA transmutation systems.

## Introduction

The Japan Atomic Energy Agency (JAEA) proposed a concept of double-strata fuel cycle, in which partitioning and transmutation are carried out in a dedicated small-scale fuel cycle attached to the commercial fuel cycle. For the dedicated transmutation system, JAEA has been proceeding with the research and development on the accelerator-driven subcritical system (ADS) cooled by lead-bismuth eutectic with a thermal output of 800 MW, whose fuel contains significant amounts of minor actinides (MA). Such reactors with large amount of MA have never been operated and examined with a critical assembly; therefore, the accuracy of MA nuclear data is concerned.

To this end, the “JAEA 800 MW ADS [1]” benchmark problem was proposed as one of problems discussed in the IAEA Co-ordinated Research Project (CRP) on “Analytical and Experimental Benchmark Analyses of Accelerator-driven Systems” [2]. The purpose of this benchmark problem is to obtain fundamental knowledge of calculation accuracy at the present time for a commercial-grade ADS. The main issues for the calculation are accuracy of nuclear data for MA and a simulation model of high-energy reactions above energies appearing in the conventional critical reactors. In the present paper, calculation results obtained in the IAEA CRP activity are shown after a description of the problem.

Such benchmark activity is one approach towards clarifying nuclear data library uncertainties through comparison with different libraries in the world. In this approach, differences due to calculation methods have to be carefully examined. Moreover, agreement of results between libraries does not always mean small uncertainty in a nuclear library, because some of the libraries contain common numerical data that can be incorrect. Another approach to clarify the uncertainty is using covariance data edited by nuclear evaluators. The covariance data enable to estimate uncertainties in reactor physics parameters, such as criticality, void reactivity and Doppler coefficient, using sensitivity coefficients. The latest version of JENDL, JENDL-4.0, contains covariance data for all nuclear data of all MA, which is one of the major updates from JENDL-3.3, which includes only a few covariance data for MA. The uncertainty estimated by using this new library is shown in the later part of the paper.

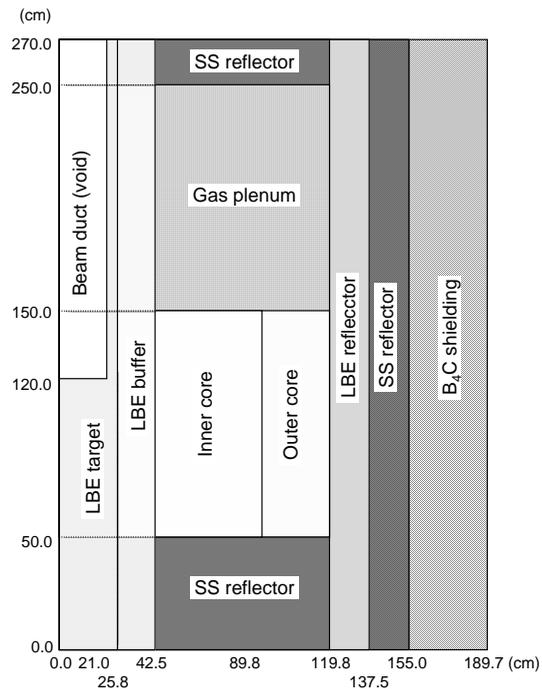
The other advantage of using covariance data and sensitivity analysis is evaluation of expected reduction of uncertainty by future critical experiments. JAEA is planning construction of the Transmutation Physics Experimental Facility (TEF-P) [3] in the second phase of the Japan Proton Accelerator Research Complex (J-PARC) project. The expected reduction by TEF-P is discussed based on the covariance data in JENDL-3.3 at the end of the paper.

## Benchmark: Description of problem

JAEA is proposing a lead-bismuth eutectic (LBE) cooled ADS. There are some advantages to a system with LBE coolant in comparison with sodium-cooled one, though thermal property is inferior to that of sodium. Chemical activity of LBE is lower than that of sodium. The neutron slowing-down power of LBE is smaller than that of sodium, and hence the neutron spectrum becomes hard in the LBE-cooled core. A hard neutron spectrum is preferable for the transmutation of MA by fission reactions. In the ADS, LBE can be used simultaneously as coolant and spallation target.

The core geometry is simplified for the benchmark calculation to a cylindrical R-Z model shown in Figure 1. Buffer assemblies are loaded between the spallation target and the fuel region to reduce the radial power-peaking factor. The buffer assembly has the same hexagonal cross-section as the fuel, but it has empty duct structure. The core is radially divided into two zones with different initial Pu loading so as to smooth the radial power distribution. Surrounding the core, LBE reflector, stainless steel shielding, and B<sub>4</sub>C shielding are loaded.

A proton linac with a proton energy of 1.5 GeV is used for the accelerator to operate the ADS. The proton beam duct is inserted along the core central axis. The axial position of beam window is 30 cm lower than the top of the active fuel region (Figure 1). A flat beam profile 40 cm in diameter is assumed in this benchmark calculation, though actually it is a Gaussian shape.

**Figure 1: R-Z Model of the ADS**


The core thermal power is 800 MW and burn-up period is 600 effective full power days (EFPD). After each burn-up cycle, all fuels are removed from the core and reloaded for the next burn-up cycle after cooling and reprocessing. However, this refuelling is not included in the benchmark requirement and burn-up is calculated only for the first 600 days.

For the core fuel, a mixture of mono-nitride of MA and plutonium (Pu) is used with a pellet density of 95%TD and a smear density of 80%TD. Zirconium-nitride (ZrN) is used with the fuel as the inert matrix. Nitrogen with  $^{15}\text{N}$  (assumed as 100% enrichment) is used for both (MA,Pu)-nitride and ZrN. The isotopic compositions of MA and Pu are based on the following assumptions: the spent PWR fuel of 45 GWd/t burn-up cooled for seven years before reprocessing and pure MA and Pu are extracted, then, MA and Pu are cooled for three years before fabricated to the ADS fuel. Further details are available in Ref. [1].

### Benchmark: Result of criticality (k-effective)

Table 1 lists the participants, codes and libraries. The TWODANT code employed by JAEA is a component of the DANTSYS code system, so JAEA and KIT used an equivalent deterministic transport code. Only CIEMAT employed a Monte Carlo code, MCNPX. Each participant used several nuclear data libraries, in which the JEFF-3.0, JEFF-3.1 and ENDF/B-VI are common. This “different codes with common library” rule enabled validation of the calculation methods of each participant, as shown in Figure 2. JAEA and CIEMAT supplied results based on ENDF/B-VI and JEFF-3.0. The results agree well by less than 0.4%dk. The results by KIT and CIEMAT based on JEFF-3.1 are rather large by 0.7%dk. However, this difference is smaller than the difference between libraries. The validity of calculation methods of each participant was confirmed by these agreements to be less than 0.7%dk.

Figure 3 shows the comparison between the JENDL, ENDF and JEFF libraries. Each library consists of the latest version and previous versions. One observation is that updates of each library result in a large difference by 2 to 3%dk in k-effective at both BOC and EOC. This means nuclear data libraries are still immature for the ADS analysis.

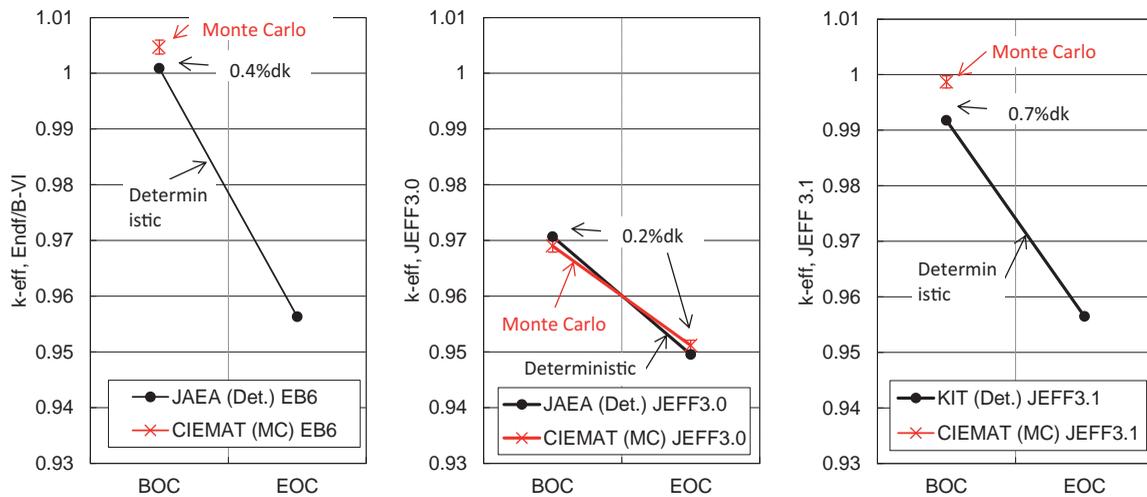
**Table 1: Participant, code and library**

Participant	Code	Library*
JAEA (Japan)	PHITS, NMTC or MCNPX (MC code for proton and neutron >20 MeV) SLAROM (cross-section code) TWODANT (deterministic neutron transport code) ORIGEN (burn-up code)	JENDL-3.3 JENDL-3.2 JENDL-4.0 ENDF/B-VI JEFF-3.0
CIEMAT (Spain)	EVOLCODE2 (MCNPX-based burn-up code)	JEFF-3.0 (JEFF-3.1) (ENDF/B-VI)
KIT (Germany)	High-energy particles are not analysed C <sup>4</sup> P, ZMIX (cross-section code) DANTSYS (deterministic neutron transport code) TRAIN (burn-up code)	ENDF/B-VII JEFF-3.1

\* Libraries in parentheses are only for the beginning of cycle (BOC).

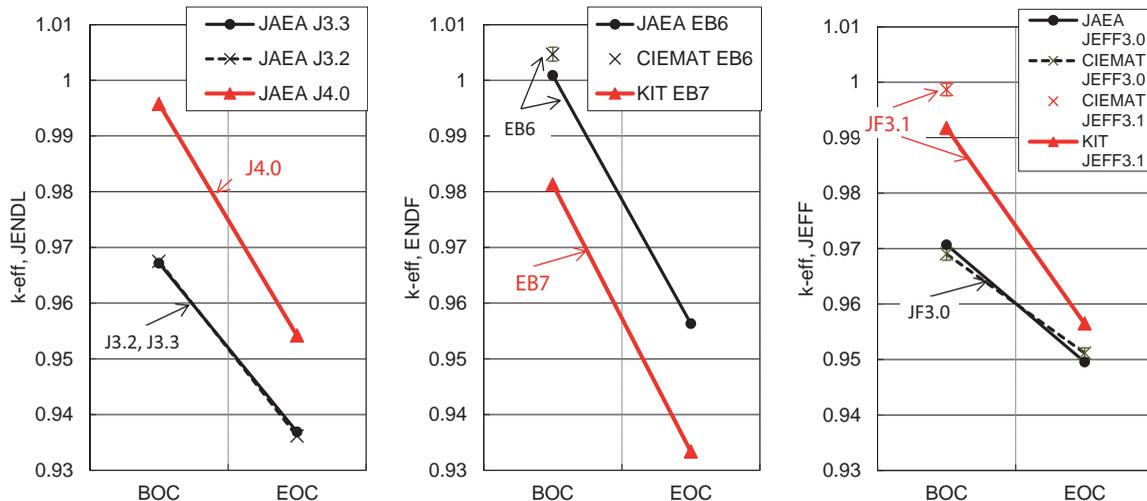
**Figure 2: Comparison based on the same library**

BOC – beginning of cycle, EOC – end of cycle



**Figure 3: Comparison of k-effective between libraries**

BOC – beginning of cycle, EOC – end of cycle



Another observation is the difference between the latest libraries by about 2%dk at the BOC. To identify responsible nuclides for this difference, authors performed additional calculations using MCNPX. The changes of k-effective were calculated when a library for one of the nuclides in the ADS system is changed to another library. The base library is JENDL-4.0, and nuclear data for one nuclide were changed to ENDF/B-VII or JEFF-3.1. As shown in Figure 4, large changes are observed for  $^{237}\text{Np}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{243}\text{Am}$ ,  $^{15}\text{N}$  and  $^{56}\text{Fe}$ . Most of these nuclides are peculiar to the ADS. Future sensitivity analyses will reveal which reaction in which energy is responsible.

The depletion of k-effective, which is defined as a decrease of k-effective from BOC to EOC and appears as slopes of lines in Figure 3, is also different among the latest libraries. It is well-known that the isomer-state branching ratio (IR) of the  $^{241}\text{Am}$  capture reaction affects depletion. A greater IR value, which means increments of  $^{242}\text{Am}$  and its daughters, results in larger depletion, because  $^{242}\text{Am}$  and its daughters have smaller reactivity than  $^{242m}\text{Am}$ , which is very fissionable. JAEA and CIEMAT employed an IR value of 80%, while KIT used 91% based on the JEFF-3.1 activation data. A recent study revealed that an IR of 85% or more is better for the fast neutrons [4]. The value in the case of the fast neutron spectrum given by JENDL-4 is around 85%.

**Figure 4:  $\Delta k_{\text{eff}}$  due to library exchange of a single nuclide from JENDL-4.0 to ENDF/B-VII and JENDL-4.0 to JEFF-3.1**

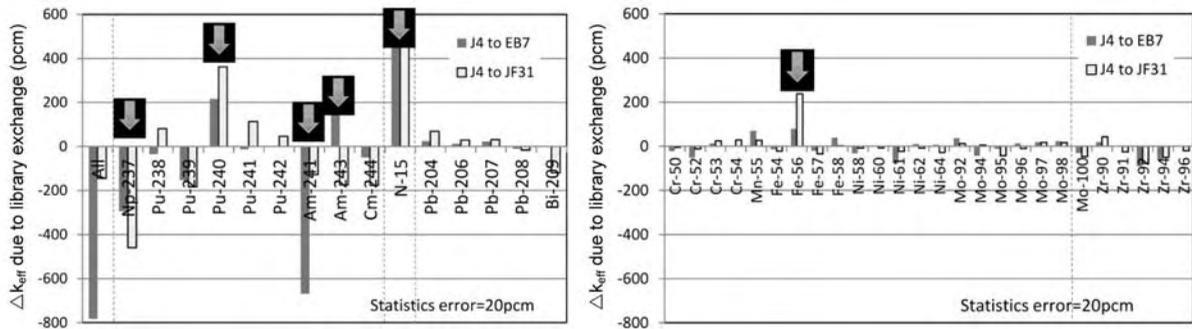
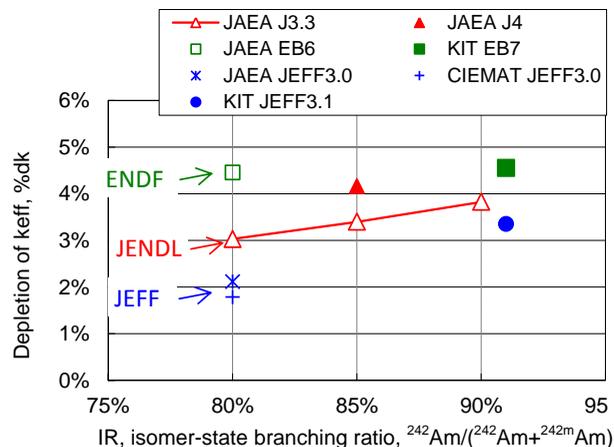


Figure 5 shows the dependency of k-effective depletion on the IR, where IR was changed only for JENDL-3.3. The results of JENDL-3.3 show the depletion of k-effective becomes larger by 0.8%dk if the IR changes from 80% to 90%. Among the three latest libraries, only the depletion of JEFF-3.1 is smaller by 1%dk than JENDL-4.0 and ENDF/B-VII, if the IR of JENDL-4.0 is compensated for up to 91%.

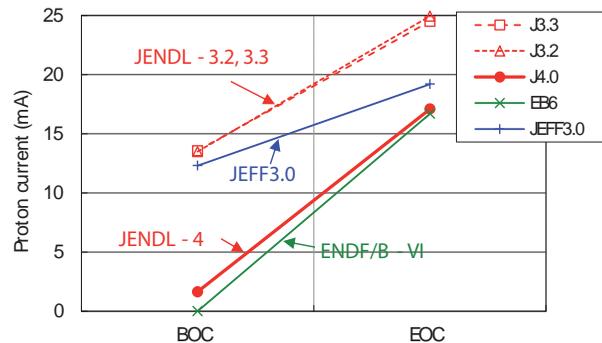
**Figure 5: Isomer-state branching ratio (IR) of  $^{241}\text{Am}$  capture reaction versus depletion of k-effective**



## Benchmark: Result of high-energy part

The calculation for the high-energy part is of importance for estimation of the proton beam current needed for pre-defined thermal output, 800 MWt. Figure 6 shows a comparison of beam current between libraries based on the JAEA result. The beam currents are much different from each other because they largely depend on criticality. Therefore, the comparison of beam current must be based on the same calculation for the low-energy part, e.g. energy less than 20 MeV.

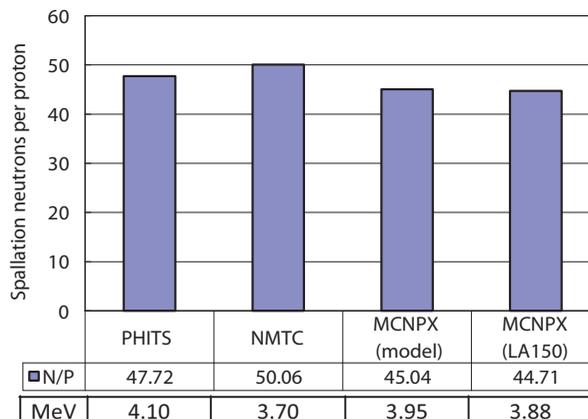
**Figure 6: Comparison of beam current between libraries based on the JAEA result**

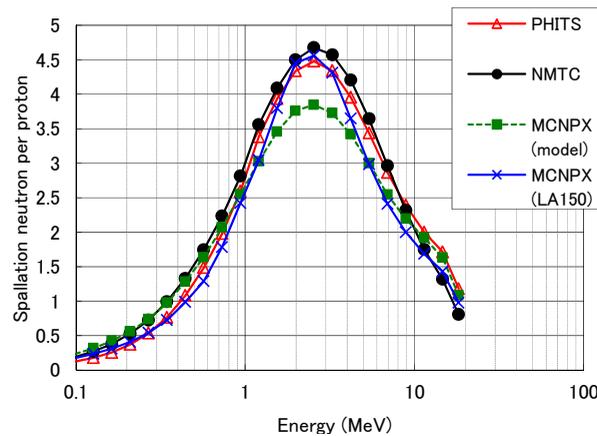
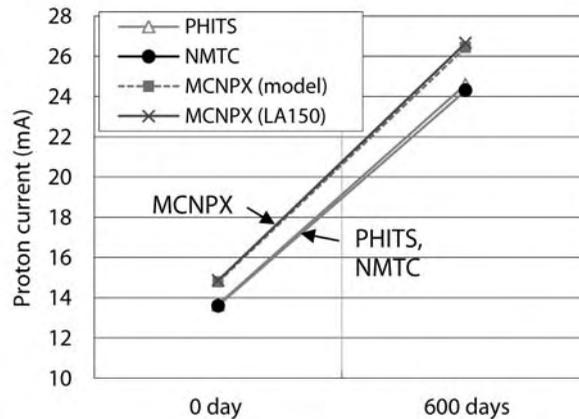


In respect to the above, the comparison between participants of the IAEA benchmark is useless because the criticality is too different. The authors therefore performed other calculations using PHITS, NMTC and MCNPX for the high-energy part, keeping the calculation of the low-energy part fixed. The NMTC code is a previous version of the PHITS code. The LA150 library or physical model was employed in the MCNPX calculation. The TWODANT code was employed for the low-energy part.

In these high-energy codes, spallation neutrons born with energy below 20 MeV or slowed down to energy below 20 MeV were cut off and stored for the low-energy code TWODANT. Figures 7 and 8 show the number, averaged energy and energy spectrum of the spallation neutron that appear at energies less than 20 MeV. The number and averaged energy of spallation neutrons by the PHITS code are larger than those of the MCNPX code. This result leads to greater effectiveness of the spallation source and weaker current in case of the PHITS and NMTC codes, as shown in Figure 9. The difference between PHITS/NMTC and MCNPXs is about 8% in the beam current. Another observation from Figure 8 is the MCNPX code using “model” and “LA-150” results in a different energy spectrum, while the averaged energy and the resulted beam current are almost same.

**Figure 7: Number of neutron and averaged energy of high-energy codes**



**Figure 8: Energy spectrum of spallation neutrons**

**Figure 9: Comparison of beam current based on the JENDL-3.3 library for the low-energy calculation**


### Benchmark: Summary

The 800 MWt ADS with significant amount of MA was analysed by the three participants for the purpose of revealing the calculation accuracy at the present time for commercial-grade ADS. The comparison based on the same nuclear data library showed the validity of the deterministic and Monte Carlo calculation methods employed by participants. Authors could thus concentrate on dependency of criticality on the nuclear data library. The following things were observed:

- Updates of each library (JENDL, ENDF and JEFF) result in a large difference by 2 to 3%dk in k-effective at both BOC and EOC. This means nuclear data libraries are still immature for ADS analysis.
- There is a difference between the latest libraries (JENDL-4.0, ENDF/B-VII and JEFF-3.1) by about 2%dk at BOC. This is due to  $^{237}\text{Np}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{243}\text{Am}$ ,  $^{15}\text{N}$  and  $^{56}\text{Fe}$ .
- The depletion of k-effective from BOC to EOC becomes larger by 0.8%dk if the isomer-state branching ratio of the  $^{241}\text{Am}$  capture reaction changes from 80% to 90%. Among the three latest libraries, the depletion by JEFF-3.1 is smaller by 1%dk than that of JENDL-4.0 and ENDF/B-VII.

As for the high-energy calculation, which is another feature of the ADS calculation, authors used three high-energy codes, PHITS, NMTC and MCNPX coupled with TWODANT, the common

low-energy code, below energies of 20 MeV. The result showed that the difference between PHITS/NMTC and MCNPX is about 8% in the beam current, that is, the current accuracy of the high-energy calculation relevant to the beam current might be around 10% at least.

Finally, the difference in the criticality by 2 to 3%dk is too large to design and construct an ADS. A more detailed analysis of the uncertainty by covariance data and its reduction by future critical experiments are discussed in the following sections.

### Evaluation of uncertainty in criticality by covariance data

The covariance data contained in the recent nuclear data libraries enable the estimation of uncertainties in reactor physics parameters, such as criticality, void reactivity and Doppler coefficient, by coupling the covariance data with the sensitivity coefficient of each cross-section to the physics parameters. The latest version of JENDL, JENDL-4.0, contains the covariance data of all nuclear data of all MA, which is one of the major updates from JENDL-3.3, which includes only few covariance data for MA (Tables 2 and 3). However, the covariance data for  $^{15}\text{N}$ , Zr, Pb and Bi are still missing in JENDL-4.0. The missing data are supplemented by those in JENDL-3.3, though most of them remain provisional data. The provisional data in JENDL-3.3 are rough data preliminarily supplied by experts. As another minor update, data concerning Fe, Cr and Ni with natural abundance are changed to those of  $^{56}\text{Fe}$ ,  $^{52}\text{Cr}$  and  $^{58}\text{Ni}$ , respectively. As to the sensitivity coefficient, the SAGEP code, based on diffusion theory and generalised perturbation theory, was employed with an 18-group structure.

**Table 2: Covariance data in JENDL-3.3**

J33 – covariance data prepared in JENDL-3.3, p – provisional covariance data (Nakagawa, 2007; Shibata, 2007)

Nuclide	Capture	Fission	$\nu$	Elastic	Inelastic	$\chi$	$\mu$ -bar
$^{238}\text{Pu}$	J33	J33	p	p	p		
$^{239}\text{Pu}$	J33	J33	J33	J33	J33	J33	J33
$^{240}\text{Pu}$	J33	J33	J33	J33	J33	J33	J33
$^{241}\text{Pu}$	J33	J33	J33	J33	J33		J33
$^{242}\text{Pu}$	J33	J33	p	p	p		
$^{237}\text{Np}$	J33	J33	J33	p	p		
$^{241}\text{Am}$	J33	J33	J33	p	p		
$^{242\text{m}}\text{Am}$	J33	J33	p	p	p		
$^{243}\text{Am}$	J33	J33	J33	p	p		
$^{242}\text{Cm}$	p	p	p	p	p		
$^{243}\text{Cm}$	p	p	p	p	p		
$^{244}\text{Cm}$	J33	J33	p	p	p		
$^{245}\text{Cm}$	p	p	p	p	p		
$^{246}\text{Cm}$	p	p	p	p	p		
$^{15}\text{N}$	p	–	–	J33	p	–	
Fe	J33	–	–	J33	J33	–	J33
Cr	J33	–	–	J33	J33	–	J33
Ni	J33	–	–	J33	J33	–	J33
$^{90}\text{Zr}$	J33	–	–	p	J33	–	
$^{91}\text{Zr}$	p	–	–	p	p	–	
$^{92}\text{Zr}$	p	–	–	p	p	–	
$^{94}\text{Zr}$	p	–	–	p	p	–	
$^{96}\text{Zr}$	p	–	–	p	p	–	
$^{204}\text{Pb}$	p	–	–	p	p	–	
$^{206}\text{Pb}$	p	–	–	p	J33	–	
$^{207}\text{Pb}$	p	–	–	p	J33	–	
$^{208}\text{Pb}$	p	–	–	p	J33	–	
$^{209}\text{Bi}$	p	–	–	p	J33	–	

**Table 3: Covariance data in JENDL-4.0**

J40 – covariance data prepared in JENDL-4.0

Nuclide	Capture	Fission	$\nu$	Elastic	Inelastic	$\chi$	$\mu$ -bar
<sup>238</sup> Pu	J40	J40	J40	J40	J40	J40	J40
<sup>239</sup> Pu	J40	J40	J40	J40	J40	J40	J40
<sup>240</sup> Pu	J40	J40	J40	J40	J40	J40	J40
<sup>241</sup> Pu	J40	J40	J40	J40	J40	J40	J40
<sup>242</sup> Pu	J40	J40	J40	J40	J40	J40	J40
<sup>237</sup> Np	J40	J40	J40	J40	J40	J40	J40
<sup>241</sup> Am	J40	J40	J40	J40	J40	J40	J40
<sup>242m</sup> Am	J40	J40	J40	J40	J40	J40	J40
<sup>243</sup> Am	J40	J40	J40	J40	J40	J40	J40
<sup>242</sup> Cm	J40	J40	J40	J40	J40	J40	J40
<sup>243</sup> Cm	J40	J40	J40	J40	J40	J40	J40
<sup>244</sup> Cm	J40	J40	J40	J40	J40	J40	J40
<sup>245</sup> Cm	J40	J40	J40	J40	J40	J40	J40
<sup>246</sup> Cm	J40	J40	J40	J40	J40	J40	J40
<sup>15</sup> N		–	–	J40		–	
<sup>56</sup> Fe	J40	–	–	J40	J40	–	J40
<sup>52</sup> Cr	J40	–	–	J40	J40	–	J40
<sup>58</sup> Ni	J40	–	–	J40	J40	–	J40
<sup>90</sup> Zr		–	–			–	
<sup>91</sup> Zr		–	–			–	
<sup>92</sup> Zr		–	–			–	
<sup>94</sup> Zr		–	–			–	
<sup>96</sup> Zr		–	–			–	
<sup>204</sup> Pb		–	–			–	
<sup>206</sup> Pb		–	–			–	
<sup>207</sup> Pb		–	–			–	
<sup>208</sup> Pb		–	–			–	
<sup>209</sup> Bi		–	–		J40	–	

Table 4 shows estimated results by JENDL-3.3 and JENDL-4.0. The coolant void reactivity is defined as a reactivity insertion when all of the coolant in the active core vanishes. The Doppler coefficient is a coefficient when temperature of all actinides uniformly increases, as usual.

**Table 4: Covariance data in JENDL-3.3 and JENDL-4.0**

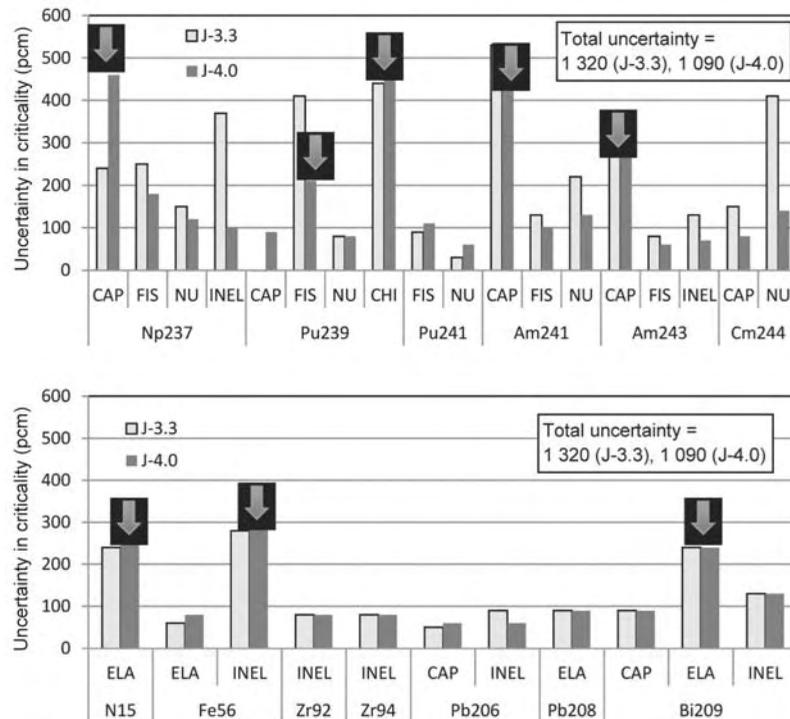
	JENDL-3.3		JENDL-4.0	
	Value	Uncertainty	Value	Uncertainty
Criticality (k-effective)	0.971	1 320 pcm (1.4%)	0.999	1 090 pcm (1.1%)
Coolant void reactivity [ $\Delta k/k$ ]	5 300 pcm	350 pcm (6.6%)	3 500 pcm	280 pcm (8.1%)
Doppler coefficient [ $T\Delta k/dT$ ]	-3.5e-4	7.0%	-3.3e-4	6.3%

### Criticality

The value is very different between JENDL-3.3 and JENDL-4.0, similarly to the result of the IAEA benchmark discussed above. The uncertainties of the two libraries, 1 320 and 1 090 pcm, are equivalent, while the breakdown is different for the actinides, as shown in Figure 10. Based on JENDL-4.0, the significant reactions for criticality are listed in Table 5. An interesting observation is that most of the nuclides listed for criticality are common to the nuclide responsible for the discrepancy of k-effective in the IAEA benchmark (Figure 4). However, the estimated uncertainty of 1 090 pcm by the covariance data is much smaller than the dispersion of k-effective among

**Table 5: Dominant reactions to uncertainty based on JENDL-4.0**

	Actinide	Others
Criticality (k-effective)	$^{237}\text{Np}$ capture, $^{239}\text{Pu}$ fission and $\chi$ , $^{241}\text{Am}$ capture, $^{243}\text{Am}$ capture	$^{15}\text{N}$ elastic, $^{56}\text{Fe}$ inelastic, $^{209}\text{Bi}$ elastic
Coolant void reactivity	$^{237}\text{Np}$ capture, $^{239}\text{Pu}$ $\chi$ , $^{241}\text{Am}$ capture	$^{12}\text{N}$ elastic, $^{56}\text{Fe}$ inelastic and $\mu$ , $^{58}\text{Ni}$ inelastic, $^{209}\text{Bi}$ elastic and inelastic
Doppler coefficient	$^{237}\text{Np}$ capture, $^{239}\text{Pu}$ $\chi$ , $^{241}\text{Am}$ capture	Zr isotopes capture, $^{206}\text{Pb}$ capture, $^{209}\text{Bi}$ capture

**Figure 10: Breakdown of criticality uncertainty**


the libraries observed in the IAEA benchmark, 2 000 to 3 000 pcm. This discrepancy of the uncertainty means that the difference in the cross-section evaluated by the individual evaluator is larger than the covariance data. The comparison of covariance data and cross-section differences among the libraries is necessary in the future work.

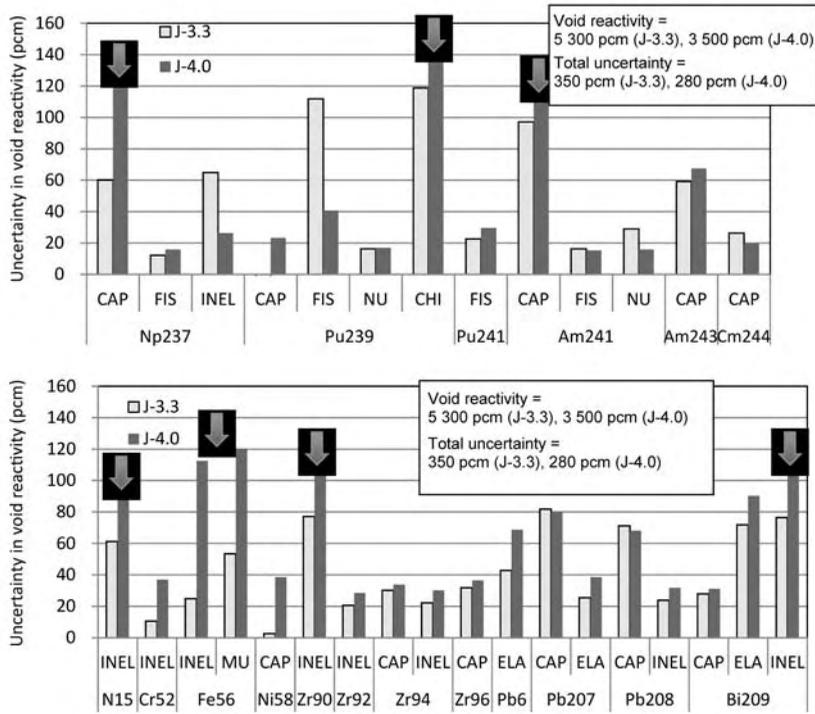
#### Void reactivity

This value is also largely different, from 5 300 to 3 500 pcm. The uncertainty established by JENDL-4.0 is 8%. The dominant reactions are almost similar to those of the criticality, as shown in Figure 11 and Table 5.

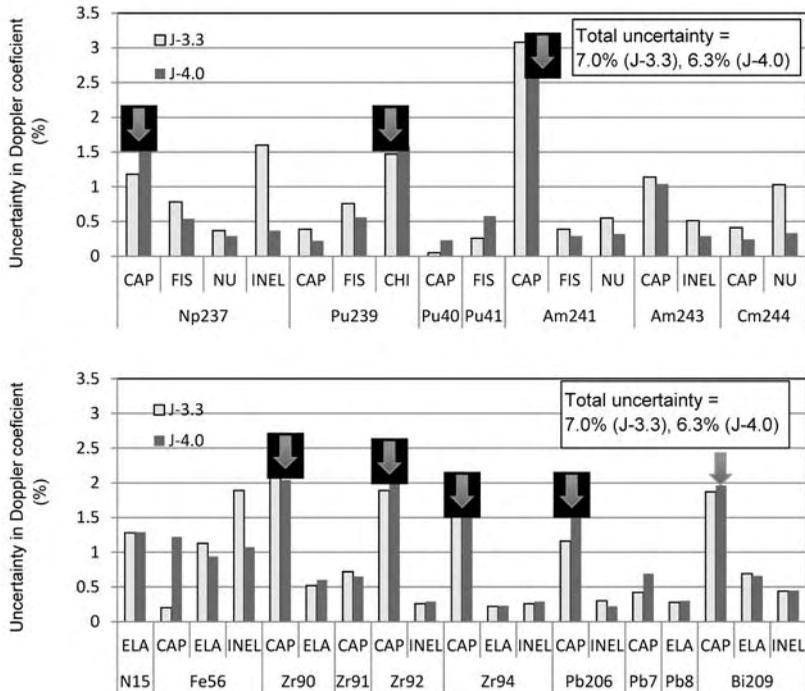
#### Doppler reactivity coefficient

The uncertainty is established by JENDL-4.0 is 6.3%. The dominant reactions among actinide are almost similar to those of the criticality, while those among other nuclides are captured by Zr isotopes,  $^{206}\text{Pb}$  and  $^{209}\text{Bi}$ .

**Figure 11: Breakdown of uncertainty of void reactivity**



**Figure 12: Breakdown of uncertainty of Doppler reactivity coefficient**



## Reduction of uncertainty in criticality by TEF-P [5]

The uncertainty estimated above can be reduced by integral experiments using critical assemblies. However, nuclides dominant to uncertainties are MA and they are very radioactive to install in the conventional critical assemblies in any significant amount. Therefore, JAEA proposes TEF-P, with a significant amount of MA in the J-PARC project. In this section, the uncertainty reduction by the TEF-P with kg-order MA is estimated.

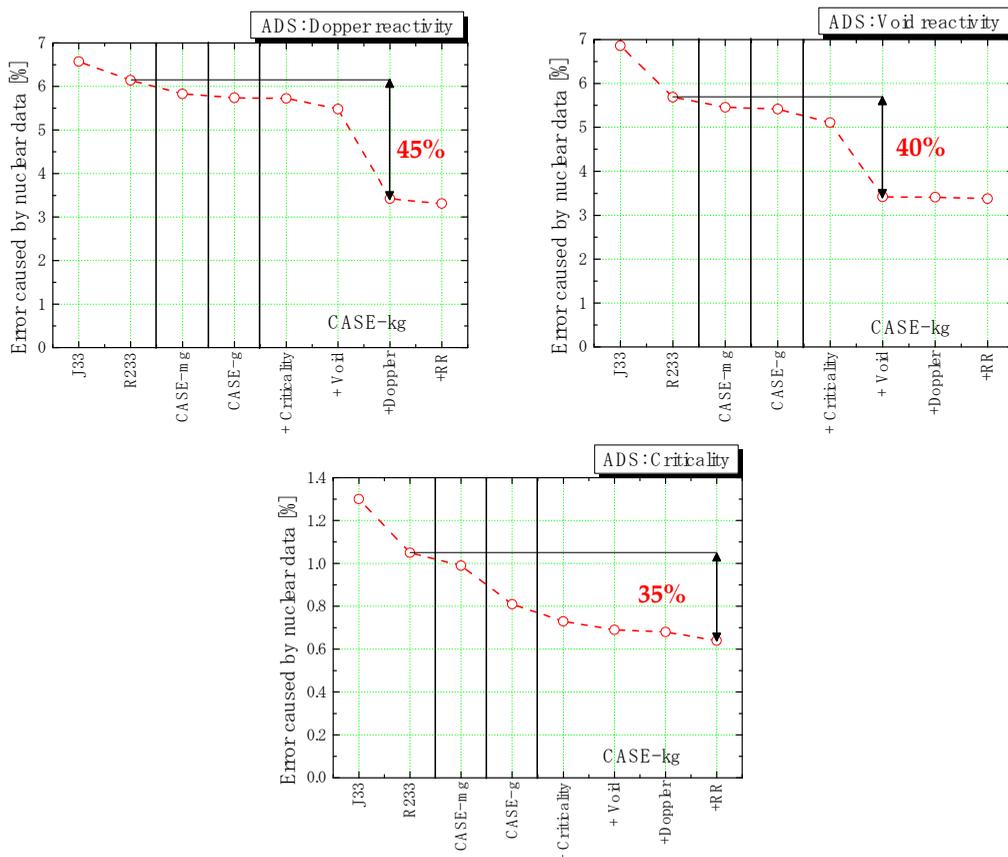
The cross-section adjustment procedure [6] was employed for the estimation of the reduction. In this procedure, the experimental data coupled with experimental and calculation error are used to adjust group cross-sections and to reduce those uncertainties. Since there is no experimental data from TEF-P, the experimental data are assumed to be equal to the calculation result, and experimental error was taken from the past experiments in the fast critical assembly (FCA) that has the same configuration as the TEF-P. Thus, the error reduction that will be brought by the TEF-P can be assessed.

Three orders of MA amount were analysed to confirm the necessity of a large amount of MA. Three orders are mg-order (CASE-mg) for chamber and foil, g-order for sample reactivity worth (CASE-g), and, kg-order (CASE-kg) for criticality, void reactivity and Doppler reactivity. The mg-order and g-order experiments can be performed in conventional critical facilities; however, the kg-order of MA is available only in next-generation facilities.

Figure 13 shows the estimated result for the uncertainty reduction, where J33 represents the original estimation by covariance data in JENDL-3.3, and R233 the reduction of uncertainty by the past 233 experiments performed in conventional facilities, such as ZPPR and FCA.

**Figure 13: Expected reduction of uncertainty by TEF-P**

J33 – original evaluation, R233 – past experiments



### Criticality

The uncertainty of the criticality can be reduced significantly in CASE-g, although the result in CASE-mg was hardly reduced. Experiments in CASE-kg are also effective for improving the error. The value was about 35% smaller than that in R233.

### Void reactivity and Doppler reactivity coefficients

The uncertainties are hardly improved in CASE-mg and CASE-g. The corresponding experiment in CASE-kg is effective, i.e. void experiment for void reactivity and Doppler experiment for Doppler reactivity coefficient. The uncertainties of void reactivity and Doppler reactivity are improved by 40% and 45%, respectively.

### Summary

The comparison among calculations for the 800 MWt ADS in the framework of the IAEA-CRP revealed that the difference of k-effective was 2-3%dk among the different nuclear data libraries. This difference is too large to design and construct an ADS.

Further investigation of the uncertainty was performed using the covariance data in JENDL-3.3 and JENDL-4.0. However, the uncertainty estimated from JENDL-4.0 was only 1 090 pcm (1.1%dk), which is much smaller than the result of the IAEA-CRP. The comparison of covariance data and cross-section differences among the libraries is necessary. The dominant reactions for uncertainty of criticality, void and Doppler reactivity were identified. The capture cross-sections of <sup>237</sup>Np, <sup>241</sup>Am and <sup>243</sup>Am were included in the dominant reactions.

Finally, the effect on uncertainty reduction of TEF-P, which is a critical facility capable of containing the kg-order MA planned in J-PARC, was assessed. The uncertainty was reduced up to 45% by the TEF-P.

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## **Session VI**

### **Transmutation System: Design, Performance and Safety**

***Chairs: P. D'Hondt, A. Stanculescu***



## Progress review of accelerator-driven system at Kyoto University Critical Assembly

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### Abstract

The Kyoto University Research Reactor Institute is going ahead with a research project on the accelerator-driven system (ADS) using the fixed field alternating gradient (FFAG) accelerator. The goal of the research project is to demonstrate the basic feasibility of ADS as an energy amplifier system using the Kyoto University Critical Assembly (KUCA) coupled with the FFAG accelerator. A series of basic ADS experiments on nuclear reactor physics has been carried out using a solid-moderated and reflected core (A-core) with 14 MeV pulsed neutrons generated by deuterium-tritium reactions. Static and kinetic experiments have been conducted for examination of the neutronic characteristics in the subcritical systems: reaction rate distribution and neutron spectrum using the foil activation method; subcriticality using several methods; neutron decay constant and neutron multiplication using the optical fibre detection systems. In the new ADS experiments using the FFAG accelerator (protons: 100 MeV energy; 5 pA intensity), the high-energy neutrons generated at a tungsten target were successfully injected into the KUCA A-core in March 2009. In March 2010, thorium-loaded ADS experiments (100 MeV energy; 30 pA intensity) were carried out to conduct a feasibility study on neutron multiplication by thorium fission reactions generated by the high-energy neutrons. In the future, on the basis of the experiments conducted in the past at KUCA relating to the minor actinides (MA) of  $^{237}\text{Np}$  and  $^{241}\text{Am}$ , ADS experiments with 150 MeV protons are planned to be carried out to investigate the MA characteristics through their reaction rate analyses, and to examine the feasibility of ADS with MA in high-energy neutron spectrum combined with several fuels (highly-enriched uranium, natural uranium and thorium) and reflectors (polyethylene, aluminium, graphite and beryllium).

## Introduction

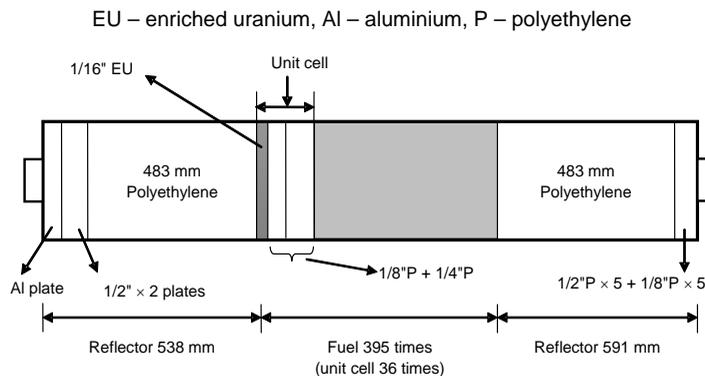
The Kyoto University Research Reactor Institute is moving ahead with a research project on the accelerator-driven system (ADS) using the fixed field alternating gradient (FFAG) accelerator [1, 2]. The goal of the research project is to establish a next-generation neutron source by introducing a synergetic system comprising a research reactor and a particle accelerator, and to demonstrate the basic feasibility of ADS as a new neutron source multiplication system using the Kyoto University Critical Assembly (KUCA) coupled with the FFAG accelerator. Prior to ADS experiments with 150 MeV protons generated from the FFAG accelerator, it was necessary to evaluate the neutronic characteristics of ADS and to establish measurement techniques for several neutronic parameters in ADS. For these purposes, a series of ADS experiments with 14 MeV neutrons [3-9] generated by the deuterium-tritium (D-T) reactions in a Cockcroft-Walton-type accelerator were carried out at the polyethylene solid-moderated and reflected core (A-core) at KUCA. Several neutronic parameters were measured in these experiments, including neutron multiplication, neutron decay constant, reaction rate distribution, neutron spectrum and subcriticality.

The world's first injection [10] of spallation neutrons generated by the high-energy proton beams into the KUCA A-core was successfully accomplished on 4 March 2009. By combining the FFAG accelerator with the A-core, the ADS experiments were carried out under the condition that the spallation neutrons were supplied to a subcritical core through proton injection (100 MeV energy; 30 Hz repetition rate; 10 pA intensity) onto a tungsten target (80 mm diameter and 10 mm thickness). In these experiments, the level of the neutron intensity generated at the tungsten target was around  $1E+06$  1/s. The objective of these experiments was to conduct a feasibility study on ADS from a reactor physics viewpoint, in order to develop an innovative nuclear reactor for a high performance transmutation system with power generation capability or a new neutron source for scientific research. Additionally, in KUCA, the thorium-loaded ADS experiments were conducted for the investigation of feasibility of neutron multiplication by thorium fission reactions obtained by high-energy neutrons through the injection of high-energy protons from the FFAG accelerator.

## KUCA core configuration

The A-core employed in the ADS experiments was essentially a thermal neutron system composed of  $^{235}\text{U}$  fuel and polyethylene-moderator and reflector. In the fuel region, a unit cell is composed of a highly-enriched uranium fuel plate  $1/16''$  thick and polyethylene plates  $1/4''$  and  $1/8''$  thick. In these ADS experiments, three types of fuel rods designated as the normal (F, see Figure 1), partial (F') and special fuel (SV) were employed. Due to safety regulations, in both ADS experiments (14 MeV neutrons and 100 MeV protons), the targets (tritium and tungsten, respectively) were located not at the centre of the core but outside the critical assembly.

**Figure 1: Side view of configuration of fuel rod (F) in the KUCA A-core**



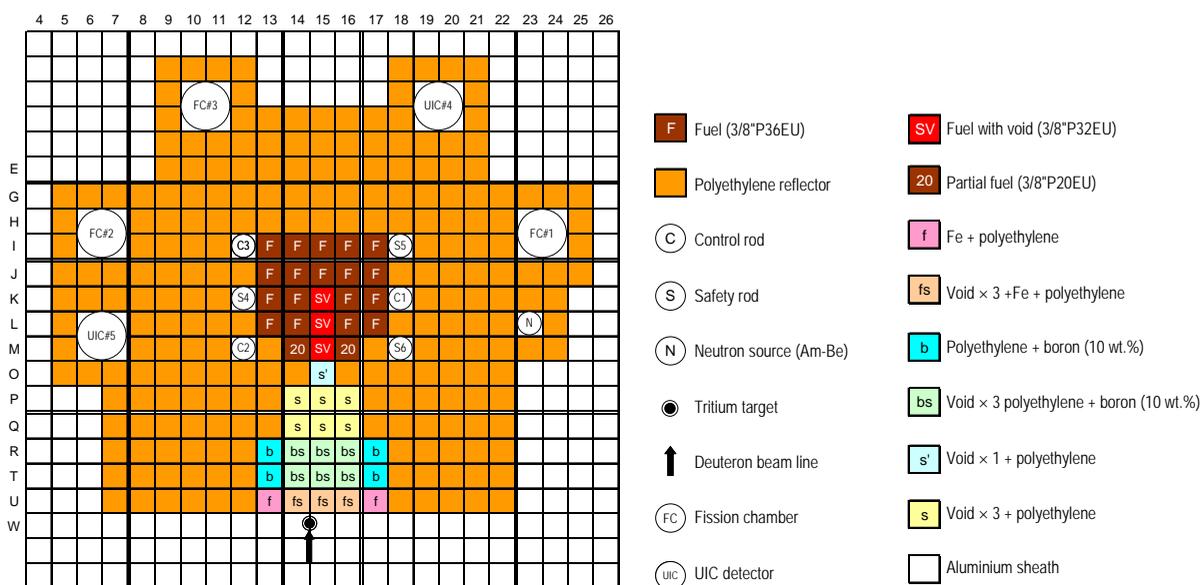
In the ADS experiments, an installation of a neutron guide and a beam duct is requisite to direct the highest number of high-energy neutrons generated from the target to the centre of the core as much as possible. The neutron guide and the beam duct were comprised by several materials inserted into the core: iron (Fe) for shielding the high-energy neutrons generated in the target region by inelastic scattering reactions; the polyethylene containing 10 wt.% boron [polyethylene + boron (10 wt.%)] for shielding the thermal neutrons moderated by absorption reactions in the reflector region; beam duct for directing collimated high-energy neutrons by streaming effect to the core region.

## ADS experiments

### ADS experiments with 14 MeV neutrons

The static and kinetic experiments (Figure 2), including the measurement of reaction rate distributions, neutron spectrum, neutron decay and subcriticality on ADS with 14 MeV neutrons were conducted at KUCA and revealed the following: measurement and calculation methods were proved reliable for the evaluation of subcriticality effects down to  $6\% \Delta k/k$ . Anticipating a conventional ADS subcriticality level around  $3\% \Delta k/k$  ( $k_{\text{eff}} = 0.97$ ), the measurement methodology and the calculation precision were considered convenient for the study of ADS at KUCA.

**Figure 2: Top view of A-core (E3 core in Figure 3) configuration with neutron guide and beam duct carried out in the ADS experiments with 14 MeV neutrons**

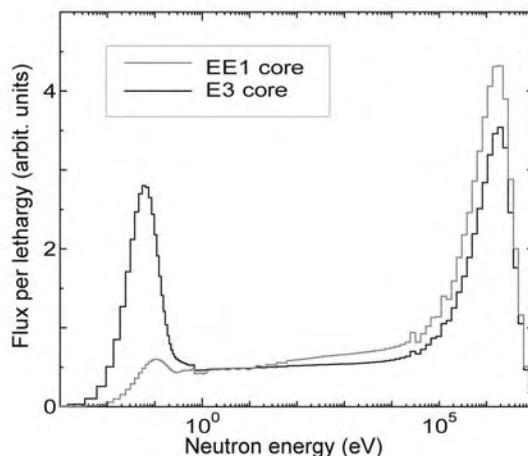


In these studies, numerical results of combined MCNP-4C3 [11] and nuclear data libraries (JENDL-3.3 [12] and ENDF/B-VI.2 [13]) showed good agreement with those of experiments, in terms of reactivity and reaction rate analyses. And, through these analyses, the foil activation method was found to be a useful measuring technique for examining the neutronic properties of ADS with 14 MeV neutrons at KUCA. For the neutron spectrum experiments using the foil activation method in the subcritical systems, C/E values in reaction rates of the experiments and the calculations with JENDL/D-99 [14] were found to have a difference of about 10%, although a large discrepancy was observed with some foils, especially in the centre of the core. However, special mention should be made of the fact that these experiments clearly revealed subcriticality dependence in reaction rate analyses.

### ADS experiments with 100 MeV protons

The ADS experiments with 100 MeV protons were carried out by varying the neutron spectrum: E3 (soft spectrum, see Figure 1) and EE1 (hard spectrum) cores shown in Figure 3. The E3 and EE1 were originally named in KUCA on the basis of the difference between cell patterns in the fuel rod.

**Figure 3: Neutron spectra in E3 (soft spectrum) and EE1 (hard spectrum) cores**



### Static experiments

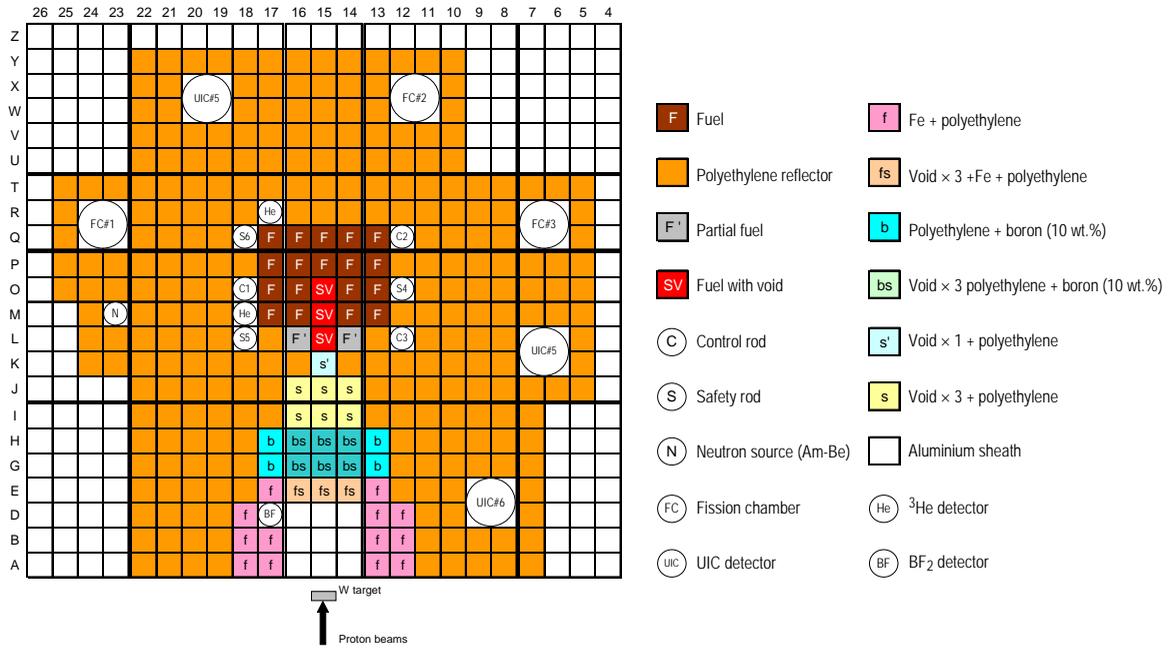
Thermal neutron flux distribution was estimated through the horizontal measurement of  $^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$  reaction rate distribution through the foil activation method using an indium (In) wire of 1.0 mm diameter. The wire was set in an aluminium guide tube, from the tungsten target to the centre of the fuel region [from the position of (13, 14 – A) to that of (13, 14 – P) in Figure 4], at the middle height of the fuel assembly. The experimental and numerical results of the reaction rates were normalised using an In foil ( $20 \times 20 \times 2 \text{ mm}^3$ ) emitted by  $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$  (threshold energy of 0.5 MeV neutrons) at the target. In this static experiment, the subcritical state ( $0.77\% \Delta k/k$ ) was also attained by the full insertion of C1, C2 and C3 rods. The numerical calculation was executed by the Monte Carlo multi-particle transport code MCNPX [15] based on the nuclear data library ENDF/B-VII [16]. The generation of spallation neutrons was included in the MCNPX calculation bombarding the tungsten target with 100 MeV proton beams. Since the reactivity effect of the In wire is considered to be not negligible, the In wire was taken into account in the simulated calculation: the reaction rates were deduced from tallies taken in the In wire setting region. The result of the source calculation was obtained after 2 000 active cycles of 100 000 histories, which led the statistical error in the reaction rates of less than 10%. The measured and calculated reaction rate distributions were compared to validate the calculation method. The calculated reaction rate distribution (Figure 5) agreed approximately with the experimental results within the statistical errors in the experiments, although these experimental errors were rather larger than those of the calculations.

### Kinetic experiments

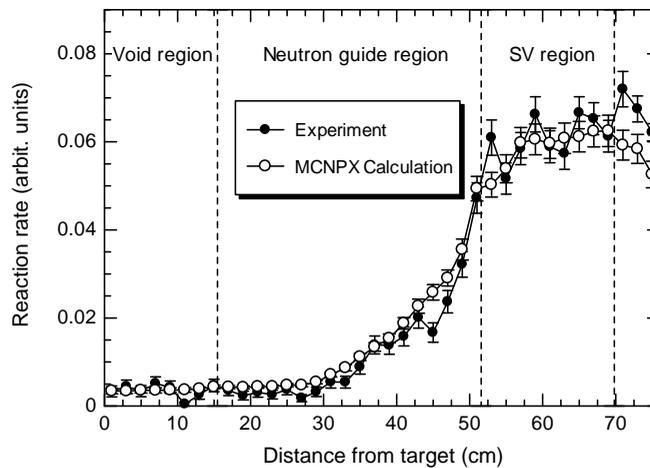
To obtain information on the detector position dependence on the prompt neutron decay measurement, the neutron detectors were set at three positions shown in Figure 4: near the tungsten target [position (17, D);  $1/2^\circ \phi$   $\text{BF}_3$  detector]; around the core [positions (18, M) and (17, R);  $1^\circ \phi$   $^3\text{He}$  detectors]. The prompt and delayed neutron behaviours (Figure 6) were experimentally confirmed by observing the time evolution of neutron density in ADS: an exponential decay behaviour and a slowly decreasing one, respectively. These behaviours clearly indicated the fact that the neutron multiplication was caused by an external source: the sustainable nuclear chain

reactions were induced in the subcritical core by the spallation neutrons through the interaction of the tungsten target and the proton beams from the FFAG accelerator. In these kinetic experiments, the subcriticality was deduced from the prompt neutron decay constant by the extrapolated area ratio method. The difference of measured results of  $0.74\% \Delta k/k$  and  $0.61\% \Delta k/k$  at the positions of (17, R) and (18, M) in Figure 4, respectively, from the experimental evaluation of  $0.77\% \Delta k/k$ , which was deduced from the combination of both the control rod worth by the rod drop method and its calibration curve by the positive period method, was within about 20%. Note that the subcritical state was attained by a full insertion of C1, C2 and C3 control rods into the core.

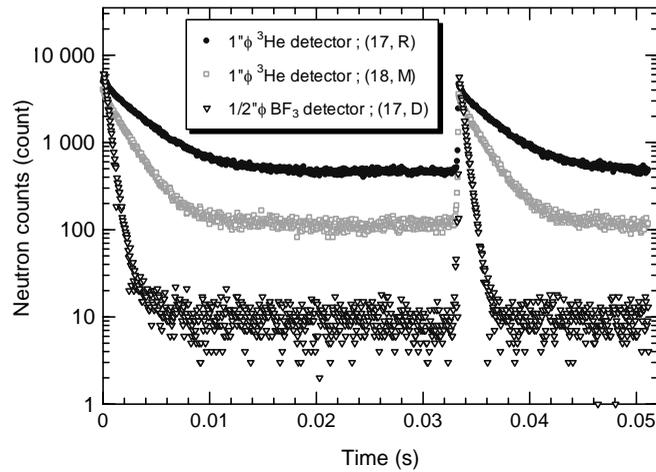
**Figure 4: Top view of configuration of A-core (E3 core) with neutron guide and beam duct carried out in the ADS experiments with 100 MeV protons**



**Figure 5: Comparison of measured and calculated reaction rate distribution from (13, 14-A) to (13, 14-P) in Figure 4**



**Figure 6: Measured prompt and delayed neutron behaviour obtained from BF<sub>3</sub> and <sup>3</sup>He detectors**

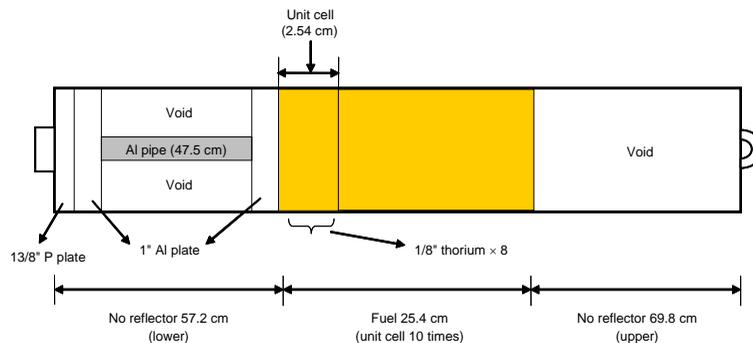


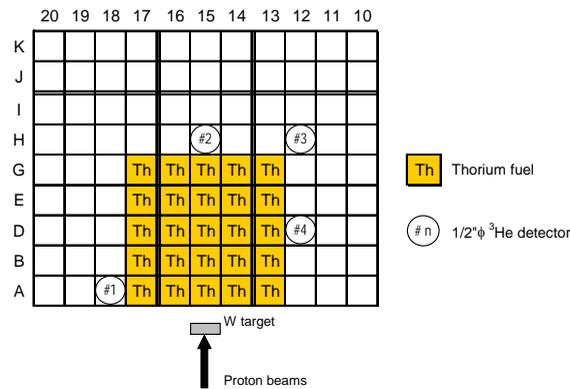
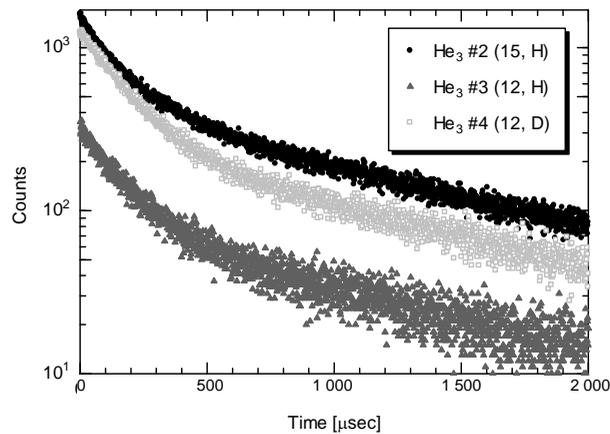
The experiments of monitoring the reactor power change were carried out for the proton beam trip and restart. In the EE1 core (hard spectrum core), the neutron signals in time series obtained by two detectors were observed in a drastic change in an interval (one minute) of beam trip and restart. The results of neutron signals revealed the tendency of a quick reduction and recovery of reactor power to the initial state during one minute. In this kinetic experiment, the subcritical state (0.69%Δk/k) was also attained by the full insertion of C1, C2 and C3 rods. As a result, the tendency of reactor power was confirmed to be almost same, even if the beam trip and restart incidents were repeated several times varying the time interval.

**Thorium-loaded ADS experiments with 100 MeV protons**

By combining the FFAG accelerator with the thorium system (Figures 7 and 8), the ADS experiments were carried out under the condition that the spallation neutrons were generated by 100 MeV protons with 30 pA intensity and 30 Hz frequency at a tungsten target. A level of the neutron intensity generated at the tungsten target was around 1E+07 1/s. The objective of these experiments was to conduct a feasibility study on the thorium-loaded ADS from the reactor physics viewpoint, i.e. as regards the confirmation of thorium fission reactions by spallation neutrons. From the results of reaction rate distribution in static experiments using <sup>115</sup>In(n,n')<sup>115m</sup>In reactions, the nuclear fission reactions by the thorium were numerically confirmed to be occurring in the core system using MCNPX (“fission turn-off” option) and ENDF/B-VII. In the kinetic experiments, the subcriticality was not deduced from the prompt neutron decay constant used in the pulsed neutron decay method, and no behaviour in the neutron decay (Figure 9) was found

**Figure 7: Side view of the configuration of fuel rod for “Th” shown in Figure 8**



**Figure 8: Core configuration of Th-loaded ADS with protons**

**Figure 9: Neutron decay behaviour at several positions in the thorium system (Figure 8)**


inside the thorium system at detector positions 2 through 4. Non-deduction was attributable to the results of very small multiplication that the numerical evaluation of the effective multiplication factor  $k_{eff}$  obtained from MCNPX based on a nuclear data library of ENDF/B-VII was  $0.03250 \pm 0.00001$  in the thorium system. In the future, experimental conditions must be improved to attain more moderated neutrons for capture reactions and the conversion ratio of capture and fission reactions needs to be examined experimentally in the thorium-loaded ADS at KUCA.

## Summary

A series of ADS experiments with 14 MeV neutrons was carried out at KUCA using the foil activation method, prior to ADS experiments with 150 MeV protons. Through these experiments, the foil activation method was found to be a useful measuring technique and the MCNP calculations with nuclear data libraries (ENDF/B-VI.2 and JENDL-3.3) were found to be reliable for examining the neutronic properties of ADS with 14 MeV neutrons.

The first ADS experiments with 100 MeV protons generated from the FFAG accelerator were successfully carried out at the KUCA A-core, although a beam commissioning in the FFAG accelerator is still under way to establish stable beam characteristics, including beam intensity and beam shaping. When stable proton beams are achieved, the actual ADS experiments with 150 MeV protons could be conducted at KUCA, such as a neutronic characteristics study on ADS with high-energy protons. Further important knowledge was attained through the thorium-loaded ADS experiments: the intensity of proton beams should be improved to be high to generate the

thorium fission reaction events in the core from the neutron multiplication viewpoint in the core, whereas the thorium fission reactions were apparently confirmed through the comparison between the experimental and numerical (MCNPX and ENDF/B-VII) analyses.

Finally, on the basis of the experiments conducted in the past at KUCA relating the minor actinides (MA) of  $^{237}\text{Np}$  and  $^{241}\text{Am}$ , the consequent ADS experiments with 150 MeV protons are planned to investigate MA characteristics through their reaction rate analyses, and to examine the feasibility of ADS with MA in high-energy neutron spectrum combined with several fuels (highly-enriched uranium, natural uranium and thorium) and reflectors (polyethylene, aluminium, graphite and beryllium).

### Acknowledgements

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## Technology readiness levels for partitioning and transmutation of minor actinides in Japan

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### Abstract

*In order to provide an assessment of the maturity of partitioning and transmutation (P&T) technology relative to its full-scale deployment, a technology readiness levels (TRL) approach was used. The definitions of TRL used in this study were based on those used in the Global Nuclear Energy Partnership (GNEP). The TRL was evaluated and the technology pathway was discussed for the systems of FBR and ADS for minor actinide (MA) transmutation, MA partitioning processes and MA-bearing fuels. The TRL for these systems/processes in Japan was found to be around TRL 3 to TRL 4. Through the evaluation, it was recognised that one of key issues was how nuclear calculation methodology for a MA-bearing core would be validated towards TRL 5. Several percentages of MA influence the core characteristics, which is important in licensing for FBR and ADS. On the other hand, a critical experiment with several kg of MA would be effective, but difficult at the existing experimental facility and would require a new one. It was also recognised that unit operations tests at engineering scale with actual spent fuel for a MA partitioning process would be difficult requirements to be satisfied at TRL 5 just after those with simulated spent fuel. For MA-bearing fuel development, engineering-scale fabrication and irradiation tests with actual separated materials were difficult requirements to be satisfied at TRL 5. The introduction of laboratory-scale tests with actual spent fuel for a MA partitioning process and with actual separated materials for MA-bearing fuel fabrication and irradiation before the engineering-scale tests (TRL 5) is an effective and efficient solution.*

## Introduction

Partitioning and transmutation (P&T) technology has been studied in Japan for more than 20 years. The long-term programme, called the OMEGA programme, was launched in 1988. The first check and review of the research and development (R&D) on P&T was made by the Japan Atomic Energy Commission (JAEC) in 2000, and the second one by the JAEC in 2008-2009; the final report was issued in April 2009 [1]. The final report covers the impact of P&T technology, the state of the art, an evaluation of the progress of the R&D, and recommendations for future R&D.

It is important for us to make a future R&D plan on P&T technology in accordance with the maturity levels of the technology, as pointed out in the final report of the second check and review. To formulate a R&D plan, the present state of the technology should be properly recognised and the present circumstances of the nuclear fuel cycle development should be understood.

The Research Committee on P&T and MA Recycle, the Atomic Energy Society of Japan (AESJ), noticed the usefulness of the technology readiness levels (TRL) approach [2] to support the planning of R&D on P&T technology. The committee decided to use the TRL approach to provide an assessment for the maturity relative to its full-scale deployment of the P&T technology in Japan. The main objective of the present TRL evaluation is not to provide absolute quantitative values of TRL, but to discuss how to conduct the research and development of P&T technology.

The present TRL evaluation was made by the Research Committee on P&T and MA Recycle, AESJ, in co-operation with the experts in each technology field. This activity was independent of the Fast Reactor Cycle System Technology Development Project (FaCT Project).

## Technology readiness levels (TRL)

TRL is a systematic metric/measurement system that supports assessments of the maturity of a particular technology from a new concept that is viable based on first principles assessment to full maturity of long-term routine operations of commercial plant. The intermediate steps are defined based on the logical progression of the research and development towards demonstration and deployment.

The TRL approach has been used on-and-off at the National Aeronautics and Space Administration (NASA) for space technology planning for many years since the 1980s. In the technology maturation process model for NASA space activities, nine levels are defined from basic principles at TRL 1 to operations at TRL 9 [2].

The TRL approach provides a relative measure of where the technology maturity is compared to the end objective of large-scale deployment. TRL is used as a programme management tool, a common understanding of science and technology exit criteria, a consistent comparison of maturity between different types of technology and a communication tool for technologists and managers.

In the Global Nuclear Energy Partnership (GNEP), the TRL approach was used and evaluated for the advanced recycling reactor, LWR spent fuel processing, waste form development, fast reactor spent fuel processing, and fuel fabrication and performance. Table 1 shows the TRL definitions for fuel fabrication used in GNEP [3]. Nine levels are defined, as with the NASA TRL model. TRL 1 to 3 are “concept development” levels, TRL 4 to 6 are “proof of principle” levels, and TRL 7 to 9 are “proof of performance” levels.

## TRL for P&T technology in Japan

The TRL definitions used in the present study were based on those in GNEP. The technologies included in the present TRL evaluation were those developed in Japan and developed within an international collaboration in which Japan participated.

**Table 1: TRL definitions for fuel fabrication used in GNEP**

TRL	Category	Generic description
9	Proof of performance	Actual fabrication facility proven through successful mission operations
8		Prototype fabrication facility completed and qualified through test and demonstration
7		System prototype demonstration in prototypic environment
6	Proof of principle	System/subsystem model or prototype demonstration in relevant environment
5		Component and/or breadboard validation in a relevant environment
4		Component and/or bench-scale validation in a laboratory environment
3	Concept development	Analytical and experimental demonstration of critical function and/or proof of concept
2		Technology concepts and/or applications formulated
1		Basic principles observed and formulated

The TRL was evaluated for the MA transmutation systems of the MA-loaded core of the fast breeder reactor (FBR) and the accelerator-driven system (ADS), and the MA recycling technology of the MA partitioning processes and the MA-bearing fuels.

### **MA-loaded core of FBR**

Table 2 shows the TRL definitions for the MA-loaded core of FBR. Although this shows only a brief description of the criteria, more detailed definitions were used for the evaluation. MA nuclear data measurements and evaluations, and integral experiments are important.

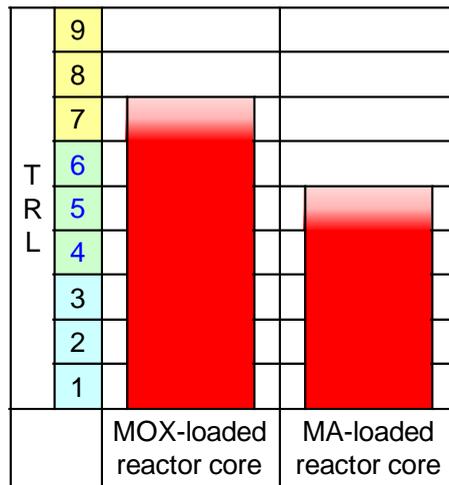
**Table 2: TRL definitions for MA-loaded core of FBR**

TRL	Category	Generic description
9 :	Proof of performance	Actual fabrication facility proven through successful mission operations :
6	Proof of principle	Nuclear design for MA-loaded core of prototype reactor
5		Critical experiment with MA Nuclear design for MA-loaded core of experimental reactor
4		MA sample irradiation Nuclear calculation code validation
3	Concept development	MA nuclear data evaluation Nuclear calculation code development
2		MA nuclear data measurement
1		Basic principles observed and formulated

Figure 1 shows the evaluated TRL for the MA-loaded core of FBR, together with the TRL for a MOX-loaded reactor core for comparison. The technology of the MA-loaded core of the FBR was evaluated as TRL 4 and is being developed at TRL 5.

Experience with a MA-loaded reactor core is limited, and even several percentages of MA loading influences the core characteristics, which is an important argument in licensing. On the other hand, a critical experiment with several kg of MA is effective, but the experiment with several kg of Am and/or Cm is difficult at the existing facility and will require a new one. One of the key issues is how the nuclear calculation methodology for the MA-loaded core would be validated to satisfy the requirements at TRL 5.

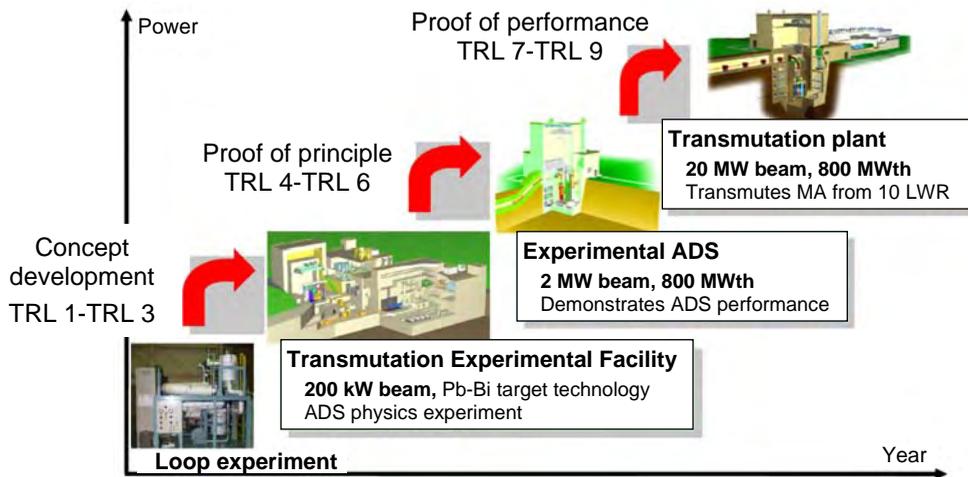
**Figure 1: TRL evaluation for MA-loaded core of FBR**



**Accelerator-driven system**

Figure 2 shows the ADS development plan from the loop experiment to the transmutation plant. The intermediate steps are the transmutation experimental facility for ADS physics experiment and the experimental ADS for demonstration of ADS performance.

**Figure 2: ADS development plan**

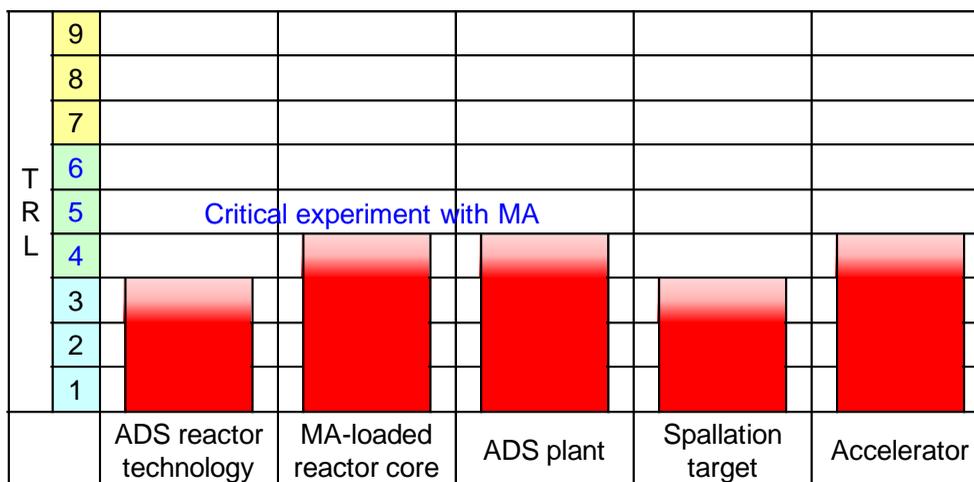


The TRL definitions for ADS are summarised in Table 3. For the development of ADS technology, several fields are related. Besides the MA-loaded reactor core, ADS reactor, plant, spallation target, and proton accelerator technology are important. To evaluate these various technologies, detailed TRL definitions for each technology were given.

Figure 3 shows the results of TRL evaluation for ADS. Technologies in several fields were evaluated. For the ADS reactor technology, basic subcritical reactor physics experiments have been completed. For the MA-loaded reactor core, the MA sample irradiation and nuclear data evaluation have been made. For the ADS plant development, lead-bismuth eutectic (LBE) loop experiments have been made. For the spallation target technology, the target irradiation tests are being made. For the accelerator, the linac has been constructed and operates at the Japan Proton Accelerator Research Complex (J-PARC).

**Table 3: TRL definitions for accelerator-driven system**

TRL	Category	Generic description
9 :	Proof of performance	Actual facility proven through successful mission operations :
6	Proof of principle	Design for MA-loaded prototype ADS
5		Critical experiment with MA; large-scale LBE loop experiment; proton beam supply to ADS test facility; spallation target mock-up
4		Nuclear calculation code validation; MA sample irradiation; beam window proton irradiation; shielding experiment/code validation
3	Concept development	Nuclear calculation code development; MA nuclear data evaluation; LBE loop experiments; target irradiation tests
2		Subcritical reactor physics; MA nuclear data measurement
1		Basic principles observed and formulated

**Figure 3: TRL evaluation for accelerator-driven system**


To develop the MA-loaded reactor core, a critical experiment with MA at TRL 5 needs a large amount of MA and a new facility. International collaboration is important and effective.

### MA partitioning processes

Table 4 shows the TRL definitions for the MA partitioning processes. The technology should be developed from the bench-scale (mg-10 g-MA), through the laboratory-scale (1-100 g-MA/day) and the engineering-scale (0.1-10 kg-MA/day), to the full-scale (1-10 kg-MA/day), using simulated and actual spent fuels.

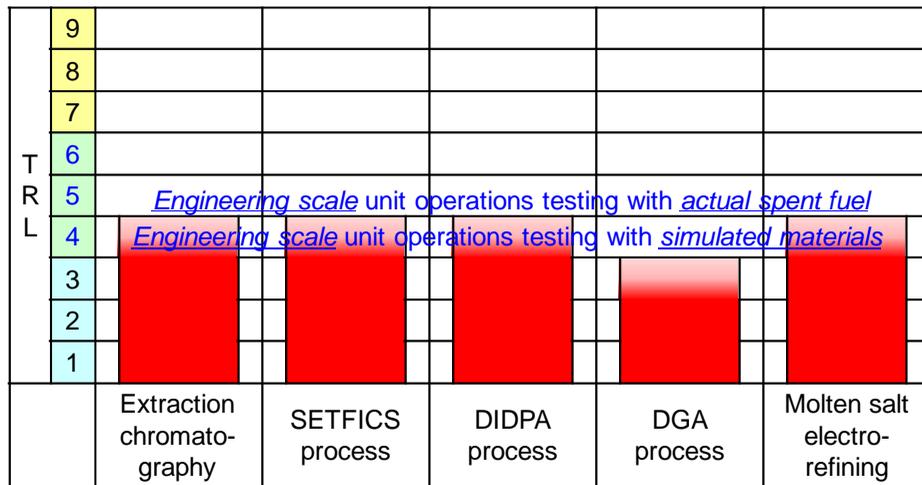
The TRL evaluated for MA partitioning processes are shown in Figure 4. Several kinds of processes have been developed for MA partitioning, and no definite difference in TRL is present among them. For extraction chromatography, the SETFICS process and the DIDPA process, bench-scale batch testing with actual spent fuel has been completed. For the DGA process, the bench-scale batch testing with simulated materials has been done. For molten salt electrorefining, the engineering-scale unit operations testing with simulated materials is under way.

To develop the MA partitioning process, the engineering-scale unit operations testing with actual spent fuel is to be completed at TRL 5. This is a difficult requirement just after those with simulated spent fuel at TRL 4.

**Table 4: TRL definitions for MA partitioning processes**

TRL	Category	Generic description
9	Proof of performance	Actual facility proven through successful mission operations
6	Proof of principle	Full-scale unit operations testing with <u>actual spent fuel</u> Process equipment design validated
5		Engineering scale unit operations testing with <u>actual spent fuel</u> Simulation models validated
4		Engineering scale unit operations testing with simulated materials Separations chemistry models developed
3	Concept development	Laboratory-scale batch testing with simulated materials Preliminary testing of equipment design concepts
2		Bench-scale batch testing with simulated materials Preliminary selection of process equipment
1		Basic principles observed and formulated

**Figure 4: TRL evaluation for MA partitioning processes**



**MA-bearing fuels**

Table 5 shows the TRL definitions for MA-bearing fuels. For MA-bearing fuel development, advances in fuel fabrication technology and fuel irradiation testing are important.

The results of the TRL evaluation for MA-bearing fuels are shown in Figure 5. Several types of MA-bearing fuels have been developed. No definite difference in TRL is present among the fuels. The fuels for the MA homogeneous recycling contain a few % of MA, while those for heterogeneous recycling contain tens % of MA. For the MA-bearing oxide and metal fuels for MA homogeneous recycling, JOYO and PHENIX irradiation tests of samples with stockpile materials have been made, respectively. For MA-bearing oxide and nitride fuels for heterogeneous recycling, characterisation of fundamental properties is being made.

To further develop MA-bearing fuels, fabrication of samples with separated (feedstock) materials is to be satisfied at TRL 5. This is not an easy requirement, and fuel development should keep in step with the MA partitioning process development. The feedstock materials will be supplied from the MA partitioning process.

**Table 5: TRL definitions for MA-bearing fuels**

TRL	Category	Generic description
9 :	Proof of performance	Actual facility proven through successful mission operations :
6	Proof of principle	Fabrication of pins with separated materials Irradiation testing of pins in prototypic environment
5		Fabrication of <u>samples</u> with <u>separated materials</u> Irradiation testing of samples in relevant environment
4		Fabrication of <u>samples</u> (rodlets, pellets) with <u>stockpile materials</u> Irradiation testing of <u>samples</u> in relevant environment
3	Concept development	Bench-scale fabrication testing with surrogates Bench-scale characterisation of fundamental properties
2		Fuel candidates selected from options based on performance data on similar systems, based on selection criteria
1		Basic principles observed and formulated

**Figure 5: TRL evaluation for MA-bearing fuels**

T R L	9				
	8				
	7				
	6				
	5	Fabrication of <u>samples</u> with <u>separated materials</u>			
	4				
	3				
	2				
	1				
		MA-bearing oxide fuel	MA-bearing metal fuel	MA-bearing oxide fuel	MA-bearing nitride fuel
	Homogeneous recycling		Heterogeneous recycling		

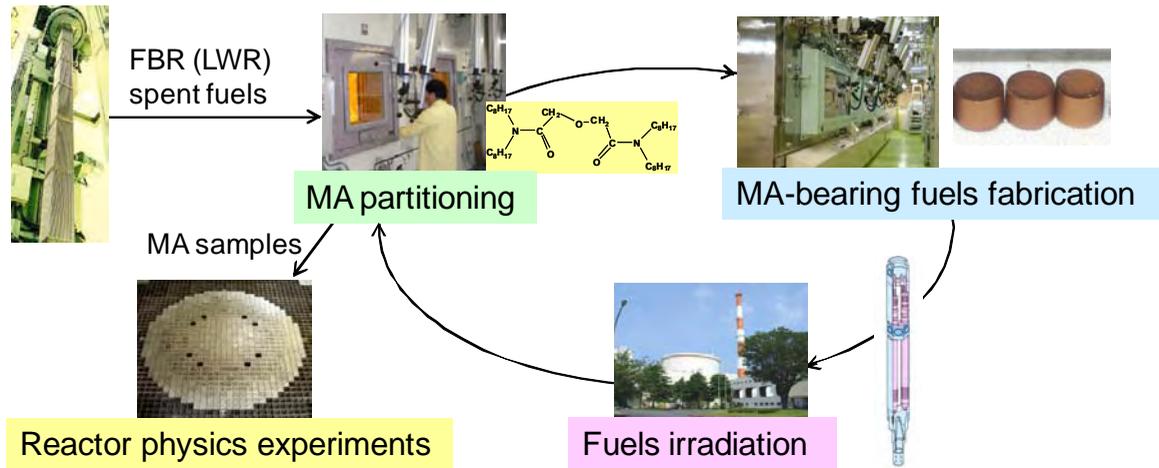
### Discussion on technology pathway with TRL

Through the TRL evaluation, it is recognised that difficult requirements are present at TRL 5 in each technology field. These include the critical experiment with a large amount of MA and a new experimental facility for ADS (and FBR), the engineering-scale unit operations tests with actual spent fuel for the MA partitioning process, and the fuel pin scale fabrication and irradiation tests with actual separated materials for MA-bearing fuels development. At TRL 5, new equipments and facilities are needed.

Before TRL 5, laboratory-scale tests with actual spent fuel should be thoroughly undertaken using existing facilities with low cost, as shown in Figure 6. Through these laboratory-scale tests, we could experience/master MA handling and treatment and compare/select partitioning processes and fuel types. Subsequently, TRL 5 could be completed with less cost and shorter schedule.

The introduction of laboratory-scale tests with actual spent fuel for MA partitioning process and with actual separated materials for MA-bearing fuel fabrication and irradiation before the engineering-scale tests (TRL 5) is an effective and efficient solution.

**Figure 6: Laboratory-scale tests with actual spent fuel and separated materials using existing facilities with low cost**



## Conclusions

The Research Committee on P&T and MA Recycle, under the Atomic Energy Society of Japan, noted the effectiveness of the TRL approach to support R&D planning for P&T technology. The TRL approach was used to provide a maturity assessment relative to full-scale deployment of P&T technology in Japan. The main objective of the present TRL evaluation was not to provide absolute quantitative values of TRL, but to discuss how to conduct the research and development of the P&T technology.

TRL was evaluated and the technology pathway was discussed for MA transmutation systems of FBR and ADS, MA partitioning processes and MA-bearing fuels. The TRL for these systems/processes in Japan was found to be around TRL 3 to 4, meaning that the P&T technology was at the level of the component and/or bench-scale validation in a laboratory environment at the most.

Through the TRL evaluation, it was recognised that difficult requirements are present at TRL 5 in each technology field and new equipments and facilities will be needed to achieve that level. For the MA partitioning process, unit operations tests at engineering scale with actual spent fuel are difficult requirements at TRL 5 just after those with simulated spent fuel. For MA-bearing fuel development, engineering-scale fabrication and irradiation tests with actual separated materials are difficult requirements at TRL 5.

The introduction of laboratory-scale tests with actual spent fuel for the MA partitioning process and with actual separated materials for MA-bearing fuel fabrication and irradiation, using existing facilities with low cost, before the engineering-scale tests at TRL 5, is an effective and efficient solution.

## Acknowledgements

The authors wish to express their thanks to the members of the task force on the TRL evaluation and the Research Committee on P&T and MA Recycle, AESJ, for their fruitful suggestions and discussion.

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## Advanced fuel cycle scenario study in the European context using different burner reactor concepts

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### Abstract

*Different types of fast spectrum dedicated burners have been proposed for the management of radioactive wastes in the frame of various advanced fuel cycle scenarios. Accelerator-driven systems (ADS) and critical low conversion ratio fast reactors have been studied, e.g. within the European context. A potential alternative system is a fusion-fission hybrid (FFH). In the present study, a sodium-cooled fast reactor driven by a D-T fusion neutron source, the subcritical advanced burner reactor (SABR) system is considered. In order to intercompare the different systems, a systematic study is under way. The performances of the two types of systems (SABR, ADS) will be compared from a minor actinide (MA) or transuranic (i.e. Pu+MA) burning potential point of view. The present paper reports preliminary results of the first phase of study, i.e. the comparison of SABR and ADS when used as minor actinides burners.*

## Introduction

According to some analysts' predictions, we witness today to a renewed interest in nuclear energy production. At present according to IAEA [1] 441 nuclear reactors are in operation (with a total power of ca. 375 GWe) and 60 more are under construction. Nuclear waste management still represents a major source of concern in public opinion [2], and partitioning and transmutation (P&T) strategies, aiming at the reduction of the burden on geological disposal (see, among many others, [3-7]) have been considered. P&T technologies, by separating and transmuted long-lived transuranic elements contained in the spent fuel, can help reduce the radiotoxicity of the wastes and in principle should also allow the reduction of the residual heat in a permanent geological repository, with a potential beneficial impact on the repository size and characteristics.

Within the framework of a collaboration between the GA-Tech (United States) and KIT (Germany), the present paper investigates the comparison between the accelerator-driven system-European facility for industrial transmutation) (ADS-EFIT) and subcritical advanced burner reactor (SABR) in a so-called "double strata" strategy developed in order to manage spent fuel (SF) inventories of European countries. The main objective of this scenario has been total waste minimisation, in a politically heterogeneous region with nation-specific nuclear energy policies. Similar studies have been performed in the past based on the use of ADS-EFIT as a transmuter [8-10]. Both ADS-EFIT and SABR offer great flexibility with respect to the choice of fuels and could also, in principle, lighten the spent fuel disposal issues in case of a future transition towards innovative energy production technologies and contribute to the management of intergenerational liabilities.

The SABR fusion-fission hybrid concept is a sodium-cooled subcritical fast reactor driven by a D-T fusion neutron source. The fusion neutron source is based on ITER physics and technology and has a maximum source strength of 500 MW<sub>th</sub>. Operating both SABR and ADS-EFIT subcritically allows to load them with a 100% transuranic (TRU) fuel, thus increasing the amount of TRU that can be burnt in the reactor, and consequently maximising the number of LWR supported by a single burner reactor. The fuel residence time in SABR is limited by the radiation damage of the cladding – not criticality – resulting in a greater single pass burning performance. A longer residence time increases fuel burn-up and facilitates reprocessing. The performances of the two types of system (SABR and ADS) will be compared when burning minor actinides (MA) or TRU (i.e. Pu+MA). The present paper reports the preliminary results of the first phase of study, i.e. the comparison of SABR and of an ADS-EFIT when used as MA burners. In practice, the performance of the two burner systems is investigated in a so-called double strata fuel cycle within a European scenario by means of COSI6 code simulations. In the scenario two groups of nations are taken into consideration: the first one with a stagnant or phasing-out nuclear policy, the second with ongoing nuclear power development. While the objective of the first group of countries is to reduce as much as possible the TRU legacy inventories by the end of the century, the objective of the second group of countries is to reprocess the spent fuel in order to recover Pu for successive use in fast reactors (FR), to reduce and successively stabilise the MA inventory, and to avoid their build-up, which could jeopardise the future deployment of fast reactors. In the scenario study both groups of countries avoid the deployment of fuel cycle facilities in isolation, but they are deploying in common a regional set of plants; regional strategies can provide in fact a framework for the implementation of innovative fuel cycle strategies with an appropriate share of efforts and facilities among different countries. The chosen scenario has already been investigated in the framework of both an EU project and of an NEA/OECD Expert Group [8], using ADS-EFIT to burn MA. In the present paper the fuel cycle scenario is analysed in terms of the required number of SABR systems to be deployed, fuel fabrication and reprocessing facilities and of the impact on some geological repository characteristics. On the basis of the European scenario the performance of the SABR system is compared to that of the ADS-EFIT. In an effort to avoid ambiguities as much as possible, the initial spent fuel inventories and other fuel cycle parameters are taken to be the same when using the two different burner reactors.

## Fast system core design used for the comparison and main results of the core neutronics assessment

In order to assess the performance of fast critical MA burner cores in the scenario analysis, both ADS-EFIT and SABR cores were simulated by the ERANOS code system [11] using the JEF-2.2 nuclear data library [12]. Both cores were loaded with the same fuel type and composition: an MgO-(TRU)<sub>2</sub> fuel having, respectively, a minor actinide-to-Pu ratio MA/Pu ~1.2 with isotopic composition coming from a mixture of LWR spent fuels, i.e. 90% of UOX cooled 30 years and 10% of MOX cooled 15 years, both with burn up 45 GWd/tHM (for isotopic composition see Table 1). The MA/Pu ratio ~1.2 is representative of TRU fuels proposed for use in ADS-EFIT [13, 14] and is mainly related to the requirement of keeping the reactivity loss during the cycle fairly constant.

**Table 1: Isotopic composition share (wt.%) of spent LWR fuel**

90% of UOX cooled 30 years and 10% of MOX cooled 15 years, both with a burn-up rate ~45 GWd/tHM (MA/Pu ~1.2)

Isotope	Content (wt.%)	Isotope	Content (wt.%)
<sup>237</sup> Np	2.2	<sup>241</sup> Am	41.0
<sup>236</sup> Pu	~0	<sup>242</sup> Am	0.1
<sup>238</sup> Pu	1.7	<sup>243</sup> Am	8.7
<sup>239</sup> Pu	21.2	<sup>242</sup> Cm	~0
<sup>240</sup> Pu	15.6	<sup>243</sup> Cm	~0
<sup>241</sup> Pu	1.8	<sup>244</sup> Cm	1.6
<sup>242</sup> Pu	5.4	<sup>245</sup> Cm	0.6

SABR [15] is a 3 000 MW<sub>th</sub> sodium-cooled subcritical fast reactor designed to burn the TRU from spent nuclear fuel. The fusion neutron source is surrounded on the outside by an annular fission core. The fusion neutron source in SABR is based on ITER physics and capable of producing up to ~500 MW of thermal power, 1.76·10<sup>20</sup> neutrons per second [16]. The annular fission core is arranged in four annular rings of fuel assemblies to facilitate a four-batch fuel cycle with a cycle time of 700 days resulting in a fuel residence time of 2 800 days, with a quarter of the fuel being removed and replaced with fresh fuel after each cycle. SABR is capable of transmuting up to 1.06 tonnes of TRU per full power year.

The fusion power necessary for this system to maintain 3 000 MW<sub>th</sub> varies between 195 MW and 325 MW at BOC and EOC respectively, and is 515 MW at beginning of fuel life (BOL).

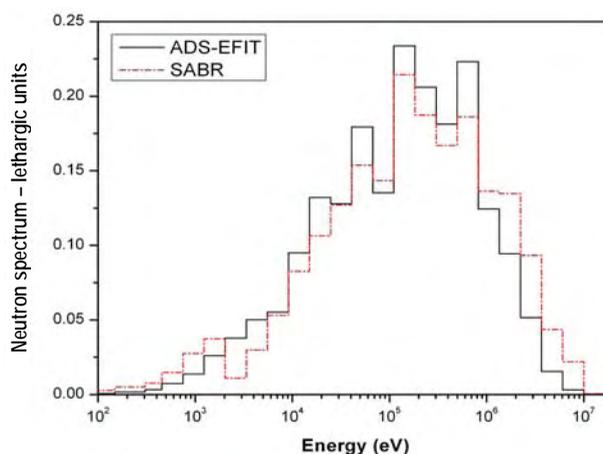
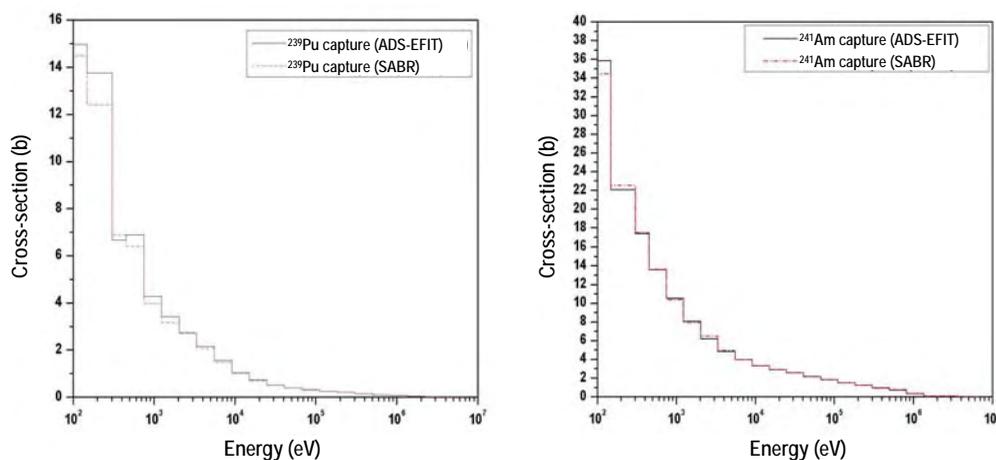
As indicated above, the strategy adopted for the ADS-EFIT is quite different, i.e. the proton beam power is kept relatively constant between BOC and EOC.

The layout of the ADS-EFIT can be found in Refs. [14, 17]. The main core parameters of SABR and of the subcritical ADS-EFIT are summarised in Table 2. The respective neutronic characteristics of SABR and of the ADS-EFIT core were investigated accounting for the different coolants (Na and Pb, respectively). The performance differences can be put in evidence by analysing the core neutron spectra and the spectrum indexes. As an example, we show in Figure 1 the comparison between the multi-group core average neutron spectra of ADS-EFIT and of the SABR. The energy spectrum differences are partly (i.e. >1 MeV) given by the different spectrum of the external neutron source and partly (i.e. <1 MeV) by the different coolants' nature, respectively, Pb and Na, and their different volume fractions (which is 63% in ADS-EFIT, and 51% in SABR). At low energy the comparison of the two neutron spectra indicates the effect on the SABR spectrum of the main <sup>23</sup>Na resonance (at ~2.85 keV), while the ADS-EFIT low-energy spectrum is determined by the neutron slowing-down in lead; the <sup>24</sup>Mg elastic scattering resonances at 19.6 keV and 83.4 keV are evident for both spectra. In particular, the two energy spectra cross each other at ~2-3 keV.

As for multi-group cross-sections, in Figure 2 <sup>239</sup>Pu and <sup>241</sup>Am capture cross-sections vs. energy (in 33 energy groups) are shown for ADS-EFIT and SABR; their differences are relatively small.

**Table 2: Main parameters of the fast reactor burners (SABR and ADS-EFIT)**

Parameter	ADS-EFIT	SABR
Fuel type	(TRU)O <sub>2</sub> -MgO	(TRU)O <sub>2</sub> -MgO
MA/Pu	~1.2	~1.2
Conversion ratio	0.0	0.0
Cycle length (EFPD)	365	700
Average fuel irradiation time (EFPD)	1 095	2 800
Average TRU content (%)	51	45
Power (GW <sub>th</sub> )	0.384	3.0
Average discharge burn-up (MWd/kg)	78	184
Reactivity loss (%Δρ)	~0.0	-60.1
Efficiency	40%	40%
TRU transmutation rate (kg/yr)	141	1 097
MA transmutation rate (kg/yr)	135	1 050
Pu transmutation rate (kg/yr)	7	46
Support ratio	5.4	42

**Figure 1: SABR and ADS-EFIT spectra comparison (at BOL)**

**Figure 2: <sup>239</sup>Pu and <sup>241</sup>Am capture cross-sections vs. energy comparison between ADS-EFIT and SABR systems**


Some ADS-EFIT and SABR spectrum indexes at the BOL and at the end of fuel life (EOL) are compared in Table 3. The values confirm, e.g. the effect of the softer spectrum of ADS-EFIT at high energy that translates e.g. in a lower  $^{238}\text{Pu}$  fission to  $^{239}\text{Pu}$  fission rate ratio.

The results of the core composition evolution during irradiation show that the specific consumption rates (referred to thermal energy produced) are very similar, as expected (Table 4). To go into detail, the results of Table 4 show that both types of cores can burn a significant amount of Am (however, a slightly lower amount is burnt by SABR per terawatt-hour-thermal because of the harder spectrum), but Cm is still building up in both cases (even if, for the average fuel residence time, the build-up is less pronounced in SABR). As for Pu, the total balance is close to zero in both cases. This is essentially due to the burn-out of  $^{239}\text{Pu}$  and the build-up of  $^{238}\text{Pu}$ . The  $^{238}\text{Pu}$  build-up has several components, but it is mostly associated with the consumption of  $^{241}\text{Am}$  due to neutron captures and successive  $\alpha$ -decay of  $^{242}\text{Cm}$ . As a consequence,  $^{238}\text{Pu}$  build-up increases with increasing  $^{241}\text{Am}$  consumption. On the other side the softer ADS-EFIT spectrum causes a higher  $^{241}\text{Am}$  capture reaction with respect to SABR, as testified by the higher amount of  $^{242\text{m}}\text{Am}$  production (Table 4). The  $^{241}\text{Am}$  fission to  $^{239}\text{Pu}$  capture cross-section ratio (Table 3) is also higher in SABR at BOL, but this trend is inverted at EOL.

In summary, these results confirm that both core types are suitable for MA legacy reduction.

**Table 3: Spectrum indexes at fuel BOL and EOL in ADS-EFIT and SABR**

		$\frac{\sigma_{c,\text{Pu-239}}}{\sigma_{f,\text{Pu-239}}}$	$\frac{\sigma_{f,\text{Pu-238}}}{\sigma_{f,\text{Pu-239}}}$	$\frac{\sigma_{c,\text{Am-241}}}{\sigma_{f,\text{Pu-239}}}$	$\frac{\sigma_{f,\text{Am-241}}}{\sigma_{f,\text{Pu-239}}}$	$\frac{\sigma_{c,\text{Am-243}}}{\sigma_{f,\text{Pu-239}}}$
ADS-EFIT	BOL	0.303	0.625	1.066	0.155	0.939
	EOL	0.320	0.612	1.099	0.147	0.974
SABR	BOL	0.350	0.545	1.046	0.176	0.903
	EOL	0.400	0.477	1.109	0.145	0.908

**Table 4: TRU consumption (kg/TWh<sub>th</sub>) of ADS-EFIT and SABR (fuel discharge values)**

Isotope	ADS-EFIT (EFPD = 1 095)	SABR (EFPD = 2 800)
$^{238}\text{Pu}$	19.3	18.2
$^{239}\text{Pu}$	-23.2	-21.9
$^{240}\text{Pu}$	-1.6	-2.3
$^{241}\text{Pu}$	0.1	1.2
$^{242}\text{Pu}$	3.4	3.0
$^{241}\text{Am}$	-45.4	-41.7
$^{242\text{m}}\text{Am}$	4.5	2.6
$^{242\text{f}}\text{Am}$	0.3	0.01
$^{243}\text{Am}$	-8.4	-6.4
$^{237}\text{Np}$	-1.2	-1.3
$^{242}\text{Cm}$	5.5	2.7
$^{243}\text{Cm}$	0.2	0.2
$^{244}\text{Cm}$	4.8	3.6
$^{245}\text{Cm}$	-0.4	0.2
$^{246}\text{Cm}$	0.1	0.0
Total Pu	-2.0	-1.8
Total MA	-40	-40.0
Total	-42	-41.8

## Transmuter type impact on double strata scenario simulation

The selected double strata fuel cycle uses UOX- and MOX-fuelled PWR in the first stratum and inert matrix (cercer) fuel (described above) in the subcritical transmuters (ADS-EFIT and SABR). The specification of the simulation scheme corresponds to a European regional scenario study which has already been investigated in the framework of the EU PATEROS project [8]. Spent UOX and MOX fuel discharged from LWR is reprocessed in order to separate TRU from fission products (FP – which, together with reprocessing losses, are sent to a geological repository). Reprocessed Pu and MA are recycled in the regional transmuter facilities and blended with TRU separated from their spent fuel (as soon as available) in subsequent cycle passes. The objective is: i) to fully reprocess the spent fuel legacy of some European countries (Group A): Belgium, Czech Republic, Germany, Spain, Sweden and Switzerland, in order to eliminate all the TRU stocks, before the end of the present century; ii) to store Pu and to stabilise the MA inventory in European countries (Group B) pursuing nuclear energy generation, as is the case in France, and planning a future deployment of fast reactors.

In an international regional fuel cycle centre ADS-EFIT or, respectively, SABR will be deployed as a common effort shared by the countries of Group A and B. The ADS-EFIT and SABR have the mission to use the plutonium of Group A (as needed by the requirement to have a Pu/MA ratio in the fuel close to one, see above) and to transmute the MA originating from two groups; transmuters burn as first choice MA coming from the Group A in order to get rid of them before the end of the century, according to scenario requirements. Plutonium of Group B is monorecycled in PWR and SF is stored for future use in a fast nuclear fleet. Transportation, economy and proliferation issues were not addressed, as they were beyond the scope of the present work.

## Scenario analysis main results and discussion

The top level goal of the scenario analyses is to eliminate the spent fuel legacy in some European countries (Group A) that do not envisage further development of nuclear energy (or even planning to phase-out) before the end of the current century. The second goal is to stabilise the MA inventory originating from the LWR operation in countries of Group B. Recovered Pu from the legacy SF inventory of Group A is used together with MA for fresh fuel fabrication to be loaded into fast transmuters.

All calculations were performed with the COSI6 code (ver. 5.1.4) [18]. A simplified scenario mass flow scheme is presented in Figure 3.

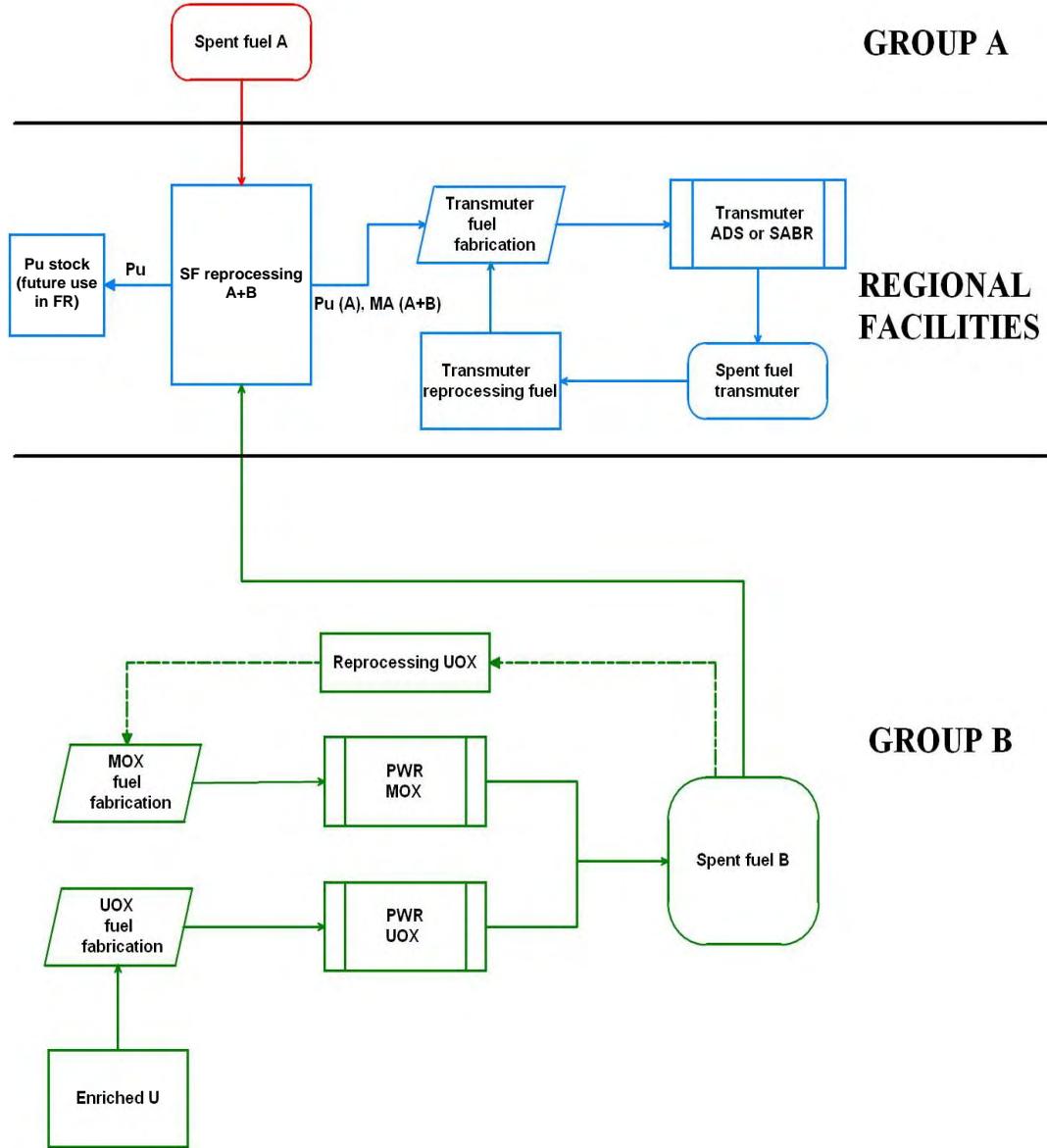
For the transmutation task ADS-EFIT and SABR transmuters were simulated in the framework of the same hypothesis and assumptions, and with the same initial and boundary conditions. Transmuters will be deployed in a regional centre, and in both cases, regional fuel cycle (FC) facilities such as a reprocessing and fuel fabrication plant for innovative fast reactor fuel, spent fuel interim storage and perhaps even a potential common repository were considered.

The annual reprocessing capacities required to manage the fuel supply are detailed in Table 5. Regional reprocessing facilities begin to operate in 2030, while transmuters' (both ADS-EFIT and SABR) start of operation is assumed to be in 2045. The legacy SF mass of Group A amounts to about 18 000 tonnes in total. The estimated German LWR UOX and MOX spent fuel inventory [19] in 2022 ca. 9 500 tonnes are added to the SF stock due to the presently planned Germany final phase-out date, thus increasing the inventory up to ca. 27 500 tonnes. The total spent fuel mass of Group A will be reprocessed by 2072 (see Figure 4), the separated plutonium is partly stored in a separated storage and partly used to feed the MA/Pu mixture to be burnt in the fast transmuters (SABR or ADS-EFIT), minor actinides (Np, Am and Cm) are collected in two separate MA stocks for each group of countries.

In general the total Pu mass available, including Pu recovered from the fuel reprocessing of the PWR 60 GWe fleet of Group B, will reach at the end of the century around 900 tonnes. This plutonium inventory is in principle sufficient to allow the transition from a full thermal reactor

**Figure 3: Simplified mass flow scheme in the regional scenario**

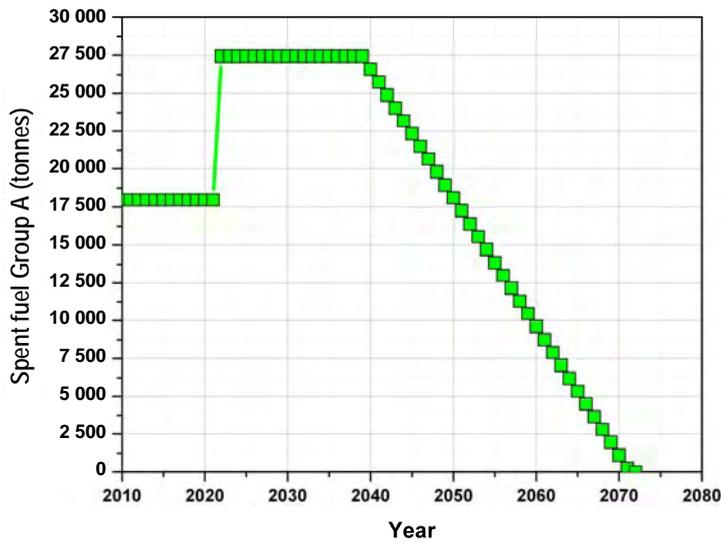
Phase I – only Groups A and B are present (until the half of the century); Phase II – the regional facility enters into operation; Phase III – MA of Group A were transmuted (before the end of the century); only Group B and regional facility remain in operation



**Table 5: Annual capacities (in tonnes) required for fuel supply with start year of operation (in parentheses)**

	Annual capacities (tonnes of heavy metal)	
	ADS-EFIT	SABR
Fuel fabrication	26 (max.)	27.5 (max.)
SF reprocessing	26 (2050)	27.5 (2050)
Reprocessing Group A –SF	850 (2040)	
Reprocessing Group B – UOX	1 500 (2010)	
Reprocessing Group B – MOX	200 (2010)	
PWR SF reprocessing (Groups A + B) – total	2 550 (2040)	

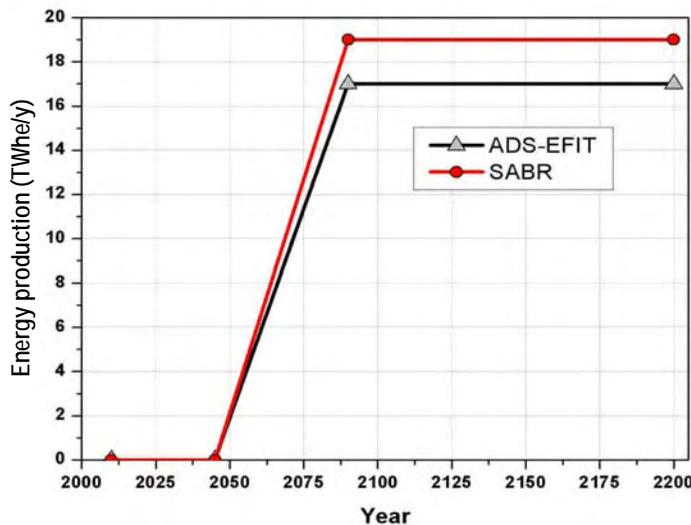
**Figure 4: Group A spent fuel mass vs. time**



to a fast reactor fleet. However the ageing of Pu (increasing <sup>241</sup>Am content) and thus the time dependent deterioration of its quality (e.g. Pu fissile content) requires further investigation on the basis of advanced modelling.

To facilitate and simplify the comparison it was assumed that both SABR and ADS-EFIT have the same net efficiency of 40%. Scenario analyses were carried out until the end of the next century, i.e. 2200, assuming the transmuters' energy production curve over the next hundred years. In COSI6 simulations the transmuter deployment pace was suitably tuned in order to meet the goals of the scenario (Figure 5).

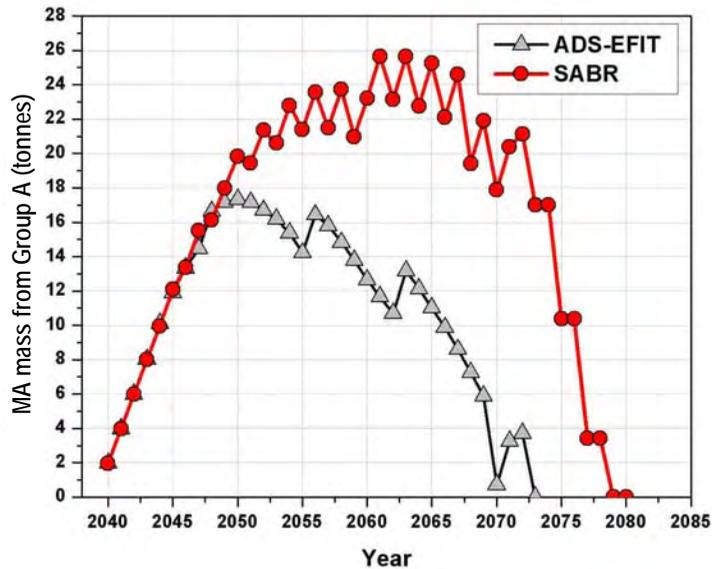
**Figure 5: Energy production for the two fast transmuter systems**



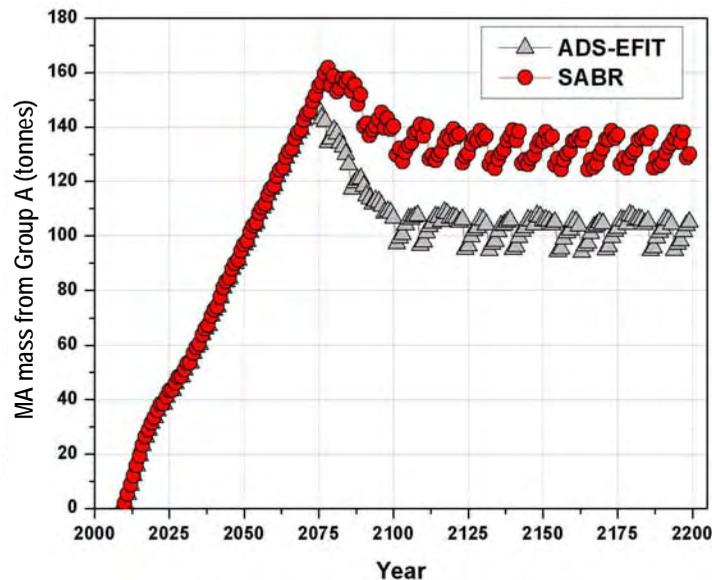
The small difference in energy production between the case with ADS-EFIT and with SABR is related to the better Am consumption rates exhibited by ADS-EFIT (see Table 4), given by the harder SABR spectrum (caused, as discussed above, by different coolant volume fractions). Moreover, it should be noted that the energy production indicated in the figure above corresponds, respectively, to that produced by ca. 2 SABR and 15 ADS-EFIT units.

The evolution with time of the content of the MA stocks of Group A and B countries are shown in Figures 6 and 7, respectively.

**Figure 6: MA mass evolution vs. time in the Group A stock**



**Figure 7: MA mass evolution vs. time in the Group B stock**



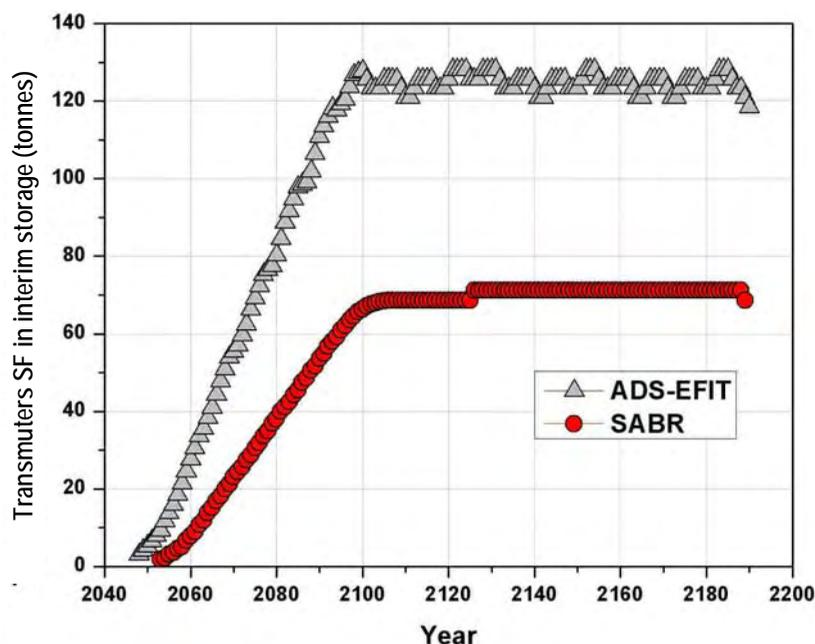
The MA stocks of Group A are eliminated ~7-8 years earlier if ADS-EFIT are used, and in both cases the goal is reached before the end of the current century, according to scenario requirements. This result follows from the discussion held above about  $^{241}\text{Am}$  consumption rate differences. As for the MA stocks of Group B, the stabilisation will be achieved at approximately the same date (i.e. around 2100). However, the stock stabilises at a higher value (~140 t) for SABR reactors than for ADS-EFIT (~100 t). This is due to the fact that for ADS-EFIT, the MA of Group B can be loaded some years earlier than is the case of SABR, as stated above. In fact, in the COSI6 simulation first MA from Group A are completely transmuted, and only after that those from Group B start to be used.

In order to give some preliminary indications of the impact of the adopted P&T strategy, some selected fuel cycle parameters such as the spent fuel heat load, neutron emission at fuel fabrication and radiotoxicity in the repository have been evaluated, and no significant differences have been found between the case with SABR reactors and the case with ADS-EFIT.

The mass of fuel in interim storage is shown in Figure 8; it can be seen that a higher mass of SF must be disposed off in the ADS-EFIT case (the ratio being roughly equal to that of transmuter burn-up, see Table 2).

The potential benefits of an international transmutation centre implementing P&T strategy are rather independent of the transmuter type.

**Figure 8: Masses of transmuter SF in interim storage vs. time**



## Conclusions

A comparison between an accelerator-driven (ADS-EFIT) and a fission-fusion hybrid (SABR) system was presented in the present paper. The systems were loaded with the same fuel composition, containing roughly 45% Pu and 55% MA. Reactor neutronics analysis proved that due to different layout, coolant nature and in particular to their volume fractions in the cores, a harder spectrum in SABR is observed, causing some minor differences in MA burning rates (especially concerning  $^{241}\text{Am}$ ) between the two systems.

Both transmuters were used for a regional scenario simulation, and their performances are rather similar, except for some differences resulting from different burning rates.

Both transmuters proved to be, in principle, valuable tools for waste management, reducing the burden on a geological disposal and allowing an easier transition to advanced fuel cycles, since the MA build-up can be controlled and stabilised.

The present paper represents a preliminary study, and further developments are foreseen in the future, in particular extending the comparison to other transmuter systems and exploring in more detail fuel cycle impacts.

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## The Multi-purpose Hybrid Research Reactor for High-tech Applications (MYRRHA)

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### Abstract

The Multi-purpose Hybrid Research Reactor for High-tech Applications (MYRRHA) is the flexible experimental accelerator-driven system (ADS) in development at SCK•CEN in replacement of the materials testing reactor BR2 of SCK•CEN in Mol (Belgium). The Belgian federal government recently approved the funding for this international project, which from 2023 onwards will contribute to the development of innovative solutions in the field of nuclear technologies.

The coupling between an accelerator, a spallation target and a subcritical core has been studied for the first time at SCK•CEN in collaboration with Ion Beam Applications (IBA, Louvain-la-Neuve) in the framework of the ADONIS project (1995-1997). ADONIS was a small irradiation facility based on the ADS concept, having a dedicated objective to produce radioisotopes for medical purposes and more particularly  $^{99}\text{Mo}$  as a fission product from highly-enriched  $^{235}\text{U}$  (HEU) fissile targets. The ad hoc scientific advisory committee recommended extending the purpose of the ADONIS machine to become a materials testing reactor (MTR) for materials and fuel research, to study the feasibility of minor actinide transmutation and to demonstrate at a reasonable power scale the principle of the ADS. The project, named MYRRHA as of 1998, then evolved to a larger installation.

MYRRHA is now conceived as a flexible irradiation facility, able to work as accelerator-driven (subcritical mode) and in critical mode. In this way, MYRRHA will allow fuel developments for innovative reactor systems, material developments for Generation IV systems, material developments for fusion reactors, radioisotope production for medical and industrial applications and industrial applications, such as Si-doping.

MYRRHA will also demonstrate the ADS full concept by coupling the three components (accelerator, spallation target and subcritical reactor) at reasonable power level to allow operation feedback, scalable to an industrial demonstrator and allow the study of efficient transmutation of high-level nuclear waste.

Since MYRRHA is based on heavy liquid metal technology (lead-bismuth eutectic), it will be able to significantly contribute to the development of lead fast reactor technology and in critical mode, MYRRHA will play the role of European Technology Pilot Plant in the roadmap for LFR.

In this paper the historical evolution of MYRRHA and the rationale behind the design choices are presented and the latest configuration of the reactor core and primary system is described.

## Introduction

Since its creation in 1952, the Belgian Nuclear Research Centre (SCK•CEN) at Mol has always been heavily involved in the conception, design, realisation and the operation of large nuclear infrastructures. The centre has played a pioneering role as regards this type of infrastructure in Europe and worldwide. SCK•CEN has successfully operated these facilities at all times thanks to the high degree of qualification and competence of its personnel and by inserting these facilities in European and international research networks, contributing to the development of crucial aspects of nuclear energy at an international level.

One of the flagships of the nuclear infrastructure of SCK•CEN is the BR2 reactor [1], a flexible irradiation facility known as a multi-purpose materials testing reactor. This reactor has been in operation since 1962 and has proven to be an excellent research tool, which has produced remarkable results for the international nuclear energy community in various fields such as materials research for fission and fusion reactors, fuel research, reactor safety, reactor technology and for the production of radioisotopes for medical and industrial applications.

BR2 has been refurbished twice, in the beginning of the 1980s and in the mid-1990s. The BR2 reactor is now licensed for operation until 2016, but around that period it will have to be decided whether another refurbishment around 2020 will have to be done or whether BR2 will be replaced by another facility. Therefore, the Belgian Nuclear Research Centre at Mol has been working for several years on the pre- and conceptual design of the Multi-purpose Hybrid Research Reactor for High-tech Applications (MYRRHA), a multi-purpose flexible irradiation facility that can replace BR2 and that is sufficiently innovative to support future-oriented research projects needed to sustain the future of the research centre.

## Scope of MYRRHA

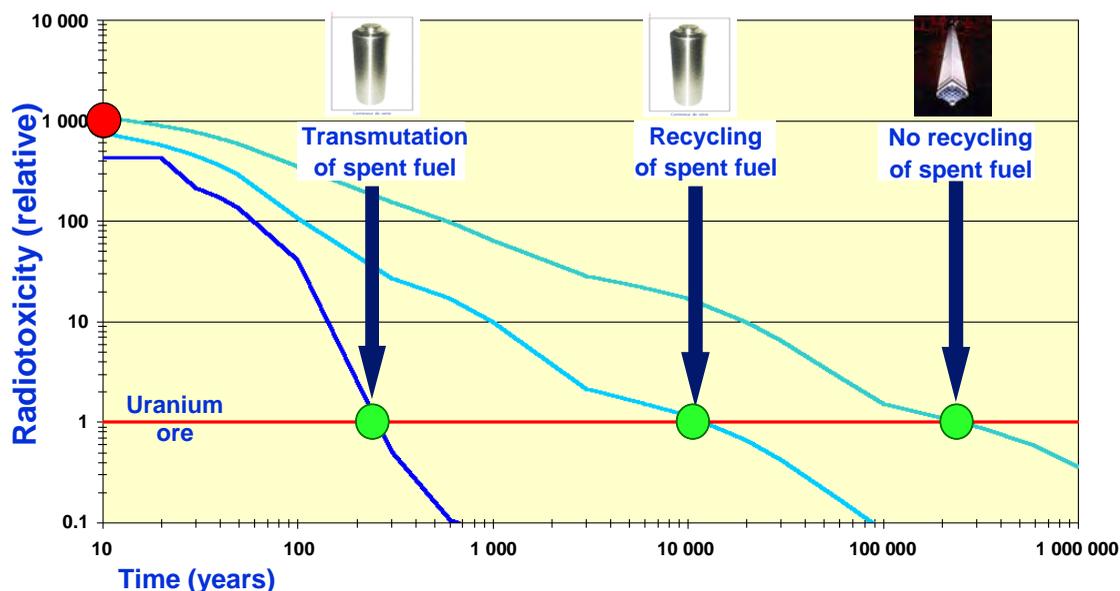
At the international level, there is a clear need to obtain a sustainable solution for the high-level, long-lived radioactive waste (HLLW) consisting of minor actinides (MA) and long-lived fission products (LLFP). These MA and LLFP stocks need to be managed in an appropriate way. Reprocessing of used fuel followed by geological disposal or direct geological disposal are today the envisaged solutions depending on national fuel cycle options and waste management policies. The required time scale for geological disposal exceeds the time span of profound historical knowledge and this creates problems of public acceptance. The partitioning and transmutation (P&T) concept (Figure 1) has been pointed out in numerous studies in the past [2-4], and more recently in the frame of the Generation IV initiative as the strategy that can relax the constraints on geological disposal and that can reduce the monitoring period to technologically manageable time scales.

The reduction of the volume and the half-life of HLLW and LLFP can be achieved conceptually by using a park of fast critical reactors that will simultaneously produce electricity and transmute the actinides, or by a “double strata” reactor park with a first stratum of reactors dedicated to electricity production using “clean fuel” containing only U and Pu and systems devoted to transmutation of HLLW and LLFP. These systems of the second stratum will be based on special fast critical reactors or more probably subcritical fast systems [accelerator-driven systems (ADS)] loaded with homogeneous fuels with a high MA content.

Even when considering a phase-out of nuclear energy, the combination of P&T and dedicated burner technologies such as ADS is needed to relax the constraints on geological disposal and reduce the monitoring period to technologically manageable time scales for existing waste. Hence, since ADS represents a possible major component in the P&T framework, the demonstration of the subcritical dedicated burner concept is needed, as indicated in the EU vision document [5] and in the strategic research agenda (SRA) [6].

Since 2000, the Generation International Forum (GIF) [7] has selected six Generation IV concepts of which three are based on fast spectrum technologies: the sodium fast reactor (SFR), the lead-cooled fast reactor (LFR) and the gas-cooled fast reactor (GFR). The SNETP community

Figure 1: Radiotoxicity as a function of time – without and with P&amp;T



has at present placed a higher priority on the SFR technology, but also indicated the need for the conception of an alternative coolant technology being lead or gas. The technological development of the fuel and materials for these concepts necessitates the availability of a flexible fast spectrum irradiation facility. The vision document and the SRA of the SNETP have also stated that Europe should be in a front-runner position for Generation IV reactor development.

Being based on the heavy liquid metal technology, lead-bismuth eutectic, MYRRHA will be able to significantly contribute to the development of lead fast reactor technology. Since Europe no longer has a fast spectrum irradiation facility, there is a clear need for a flexible fast spectrum irradiation facility in support of the development of the different fast reactor systems: SFR, LFR and GFR. As it will also be operated in critical mode, MYRRHA can even better play the role of European Technology Pilot Plant in the roadmap for LFR.

### Objectives of MYRRHA

With the scope described in the previous section, MYRRHA should therefore target the following applications catalogue:

- To demonstrate the ADS full concept by coupling the three components (accelerator, spallation target and subcritical reactor) at reasonable power level to allow operation feedback, scalable to an industrial demonstrator.
- To allow the study of efficient transmutation of high-level nuclear waste, in particular minor actinides that would request high fast flux intensity ( $\Phi_{>0.75\text{MeV}} = 10^{15} \text{ n/cm}^2\text{s}$ ).
- To be operated as a flexible fast spectrum irradiation facility allowing for:
  - Fuel developments for innovative reactor systems, which need irradiation rigs with a representative flux spectrum, a representative irradiation temperature and high total flux levels ( $\Phi_{\text{tot}} = 5 \times 10^{14} \text{ to } 10^{15} \text{ n/cm}^2\text{s}$ ); the main target will be fast spectrum Generation IV systems which require fast spectrum conditions.
  - Material developments for Generation IV systems, which need large irradiation volumes ( $3000 \text{ cm}^3$ ) with high uniform fast flux level ( $\Phi_{>1\text{MeV}} = 1\text{--}5 \cdot 10^{14} \text{ n/cm}^2\text{s}$ ) in various irradiation positions, representative irradiation temperature and

representative neutron spectrum conditions; the main target will be fast spectrum Generation IV systems.

- Material developments for fusion reactors which need also large irradiation volumes ( $3\,000\text{ cm}^3$ ) with high constant fast flux level ( $\Phi_{>1\text{MeV}} = 1\sim 5\cdot 10^{14}\text{ n/cm}^2\text{s}$ ), a representative irradiation temperature and a representative ratio approximately  $\text{He/dpa(Fe)} = 10$ .
- Radioisotope production for medical and industrial applications by:
  - holding a back-up role for classical medical radioisotopes;
  - focusing on R&D and production of radioisotopes requesting very high thermal flux levels ( $\Phi_{\text{th}} = 2\text{ to }3\cdot 10^{15}\text{ n/cm}^2\text{s}$ ) due to double capture reactions.
- Industrial applications such as Si-doping which need a thermal flux level depending on the desired irradiation time: for a flux level  $\Phi_{\text{th}} = 10^{13}\text{ n/cm}^2\text{s}$  an irradiation time of the order of days is needed and for a flux level of  $\Phi_{\text{th}} = 10^{14}\text{ n/cm}^2\text{s}$  an irradiation time of the order of hours is needed to obtain the required specifications.
- The demonstration of LBE technology.
- To contribute to the demonstration of LFR technology and the critical mode operation of heavy liquid metal cooled reactor as an alternative technology to SFR.

## Genesis of MYRRHA and its evolution

### *The ADONIS project (1995-1997)*

The coupling between an accelerator, a spallation target and a subcritical core has been studied for the first time at SCK•CEN in collaboration with Ion Beam Applications (IBA, Louvain-la-Neuve) in the framework of the ADONIS project (1995-1997). ADONIS was a small irradiation facility, based on the ADS concept, having a dedicated objective to produce radioisotopes for medical purposes and more particularly  $^{99}\text{Mo}$  as a fission product from highly-enriched  $^{235}\text{U}$  fissile targets. The proposed design was of limited size with an accelerator of 150 MeV and a core with a power of around  $1.5\text{ MW}_{\text{th}}$ . The subcritical core was made of the  $^{235}\text{U}$  targets for production of the  $^{99}\text{Mo}$  without other driver fuel. The system was a thermal spectrum machine and therefore water was used as coolant and moderator.

### *From ADONIS to MYRRHA (1998-2005)*

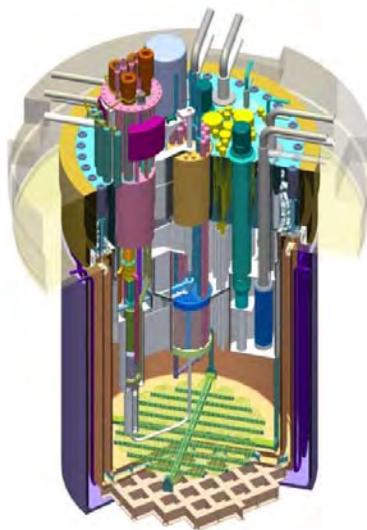
The ad hoc scientific advisory committee recommended extending the purpose of the ADONIS machine to become a materials testing reactor for material and fuel research, to study the feasibility of transmutation of the minor actinides and to demonstrate at a reasonable power scale the principle of the ADS.

This decision was taken around the same time of the last BR2 refurbishment. It was then clear that a subsequent BR2 refurbishment would be compulsory around 2020. Even if such refurbishment remains technically possible, a new machine would be better adapted to the new needs, in particular for Generation IV and fusion research. The project, named MYRRHA as of 1998, has evolved to a larger installation.

In mid-2002, a first pre-design file of MYRRHA, the “MYRRHA Draft – 1” file [8] with a core nominal power of  $30\text{ MW}_{\text{th}}$ , was submitted to an International Technical Guidance Committee for review. This international panel consisted of experts from research reactor designers, ADS development, reactor safety authorities and spallation target specialists. No show stopper was identified in the project, but some recommendations were made. The design was upgraded and the MYRRHA project team has favoured as much as possible mature or less demanding technologies in terms of research and development. In its 2005 version, MYRRHA consisted of a linear proton accelerator delivering a  $350\text{ MeV} \times 5\text{ mA}$  beam to a windowless liquid Pb-Bi spallation

target that in turn couples to a Pb-Bi-cooled, subcritical fast core of 50 MW thermal power. The so-called “Draft – 2” design (Figure 2), published in early 2005 [9], is summarised in Ref. [10], for which a business plan was also written in 2007 and is updated on a regular basis [11].

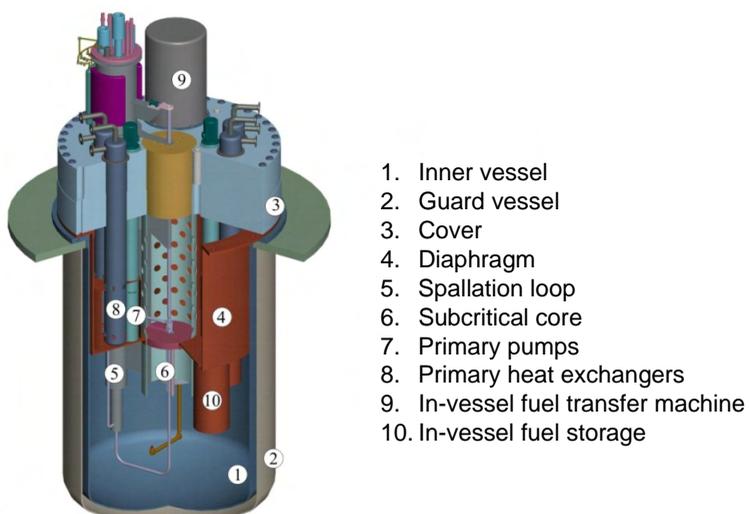
**Figure 2: MYRRHA – Draft 2**



### **From MYRRHA to XT-ADS (2005-2009)**

SCK•CEN offered to use the MYRRHA 2005 design file as a starting basis for the Experimental Facility Demonstrating the Technical Feasibility of Transmutation in an Accelerator-driven System (XT-ADS) design in the FP6 EUROTRANS Integrated Project. This allowed optimising an existing design towards the needs of XT-ADS and within the limits of the safety requirements instead of starting from a blank page. MYRRHA and XT-ADS differ however on several points such as the accelerator (linac, 600 MeV beam instead of 350 MeV), temperature level, plutonium vector in the fuel, fuel pitch and the simplification of several major components resulting in a lower power density core, an increased core power of 57 MW and a neutron flux of  $7 \times 10^{14}$  n/cm<sup>2</sup>s instead of  $1 \times 10^{15}$  n/cm<sup>2</sup>s (>0.75 MeV).

**Figure 3: XT-ADS**



### ***From XT-ADS to FASTER (2009-2011)***

The performances of XT-ADS do not fully correspond to the objectives described in the previous section. Therefore, it is clear that future design work is needed to bring this design in line with the objectives and applications catalogue.

First of all, since the objective of MYRRHA is to operate both in a subcritical mode and a critical mode, an analysis will be performed to establish to what extent the design of XT-ADS (which only considers subcritical mode operation) needs to be modified to respond to the objective to operate in critical mode. In this respect, it is clear that reactor control and scram systems have to be included in the design. Working in critical mode will significantly modify the safety characteristics of the facility and the impact of the safety feedback on the design needs to be taken into account.

A second major topic for the update of the MYRRHA design is to obtain the targets set in the applications catalogue. The XT-ADS concept is able to reach the different targets in terms of irradiation conditions listed in the applications catalogue except for the targeted fast flux level above  $E = 0.75$  MeV of  $10^{15}$  n/cm<sup>2</sup>s. In XT-ADS, this flux level reaches  $\Phi_{>0.75\text{MeV}} = 0.7 \times 10^{15}$  n/cm<sup>2</sup>s. To increase this flux level, it is obvious that the total power and power density will have to be increased. Therefore, already a preliminary study [12] was performed to investigate what would be the most optimal design changes to reach this target of  $\Phi_{>0.75\text{MeV}} = 10^{15}$  n/cm<sup>2</sup>s given the different design constraints. Preliminary conclusions of this study resulted in preserving the existing design choices of XT-ADS except for:

- an increase of the power from 57 MW<sub>th</sub> to 85 MW<sub>th</sub>;
- an increase of the power density by decreasing the number of fuel assemblies from 72 to 64 for the equilibrium core;
- an increase of the allowable temperature increment over the core from 100°C to 130°C, resulting in a decrease of the inlet temperature from 300°C to 270°C (the outlet temperature remains at 400°C);
- an increase of the beam current from 2.2 mA to 3.0 mA for ADS operation (during normal operation);
- an increase of the Pu content from 33% in a depleted uranium matrix to 35% in a natural uranium matrix.

The study pointed out that the implications of these modifications can be accommodated by the design, but a detailed confirmation is still needed. These aspects for the upgrade of the MYRRHA design, now called FASTER, along with all progress towards an advanced engineering design level for all components have been integrated in the work programme of the Central Design Team (CDT) within the FP7 project.

### **Current FASTER specifications and design**

The XT-ADS design choices made in the IP-EUROTRANS project have been reviewed in view of the different objectives set forward for FASTER and outlined in the previous sections. This process, undertaken during the first year of the CDT project, ended with the definition of the specifications and of individuation of XT-ADS design changes to arrive at a FASTER configuration able to fulfil the given objectives. FASTER shall be a flexible facility able to accommodate both critical and subcritical core configurations to be defined according to the needs of the experiments to be performed.

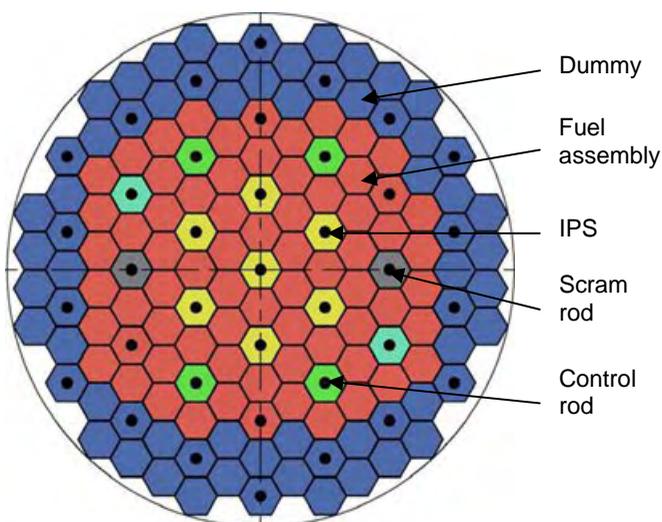
The primary and secondary systems shall be designed to evacuate a maximum thermal power of 100 MW (20 MW more than XT-ADS). This gives the required flexibility for the core power configurations while limiting the facility size and cost. The main components/systems (primary pumps, heat exchangers, secondary system and safety decay heat removal system)

shall be of the same XT-ADS type, though with increased size. All the FASTEF components are optimised for the extensive use of the remote handling system during components replacement, inspections and handling.

We are analysing the feasibility of a window-type, centred spallation target with a proton beam that impinges directly into the primary coolant. This solution could generate a great simplification in the primary system, as more space will be made available inside the reactor vessel to accommodate the in-vessel components, it also will make the core support plate design simpler, and a dedicated system to evacuate the target power (primary coolant driven by the primary pumps perform this function) would not be needed.

The fuel assemblies' (FA) size is slightly increased so as to accommodate the window target in the core centre. Consequently the in-pile test sections (IPS), which will be located in a dedicated FA position, are larger in diameter and will allow greater flexibility for experiments. Thirty-seven FA positions can be occupied by IPS or by the target (the central one for core in subcritical configuration) or by control and shutdown rods (in the core critical configuration). This will not only allow greater flexibility in the choice of the more suitable position (neutron flux) for each experiment but will simplify the design of the primary system. Figure 4 shows a horizontal cut inside the core.

**Figure 4: Horizontal cut in the FASTEF core**



The requested high fast flux intensity shall be obtained optimising the core configuration geometry (fuel rod diameter and pitch) and maximising the power density (the possibility to manufacture fuel with enrichment up to 35% is still under investigation). We are evaluating, for the first core loadings, the use of 15-15Ti as cladding material instead of T91. Thanks to the use of LBE as coolant, the core inlet operating temperature will be lower (down to 270°C), decreasing the risk of corrosion and allowing to increase the core  $\Delta T$  that together with the adoption of reliable and passive shutdown systems will permit to meet the high fast flux intensity target.

### Project schedule and budget

During the period 2010-2014, the following tasks will be accomplished in parallel:

- The detailed engineering design part of the design work will be performed through the FP7 project CDT, as described in the previous section, and will continue until 2014.
- Drafting of the technical specifications of the different procurement packages, the publication of the call for tenders, the comparison of the technical and financial proposals from the contractors and finally the awarding of the manufacturing contracts (2014-2015).

- Development and testing of key innovative components (for the accelerator and for the reactor) will run in parallel in the same time slot.
- Licensing and permitting activities, namely the writing of a Preliminary Safety Assessment Report, the Environment Impact Assessment Report and the Preliminary Dismantling Plan, the objective being to obtain the licensing permit and permit for construction at the end of 2014.

The construction period of the components and the civil engineering work will take place over a three-year period (2016-2018) followed by a one-year period for assembling the different components in 2019. The commissioning at progressive levels of power will be completed over a three-year period (2020-2022) with the final objective of being at full power operation in 2023.

In the design and licensing of MYRRHA both modes of operation are foreseen from the beginning. A minimum three-year preparatory period is considered necessary for the progressive commissioning of the facility which will include both operation modes.

To obtain an answer concerning the viability of the ADS concept in an international context as soon as possible and thereby to allow evaluating the realism of subcritical burners in a double strata fuel cycle approach, it was opted to start in MYRRHA with the demonstration of ADS and hence the full power coupling with an accelerator. The MYRRHA facility is thus projected to be initially operated as an ADS in subcritical mode for the demonstration of the full power ADS operation and the efficient transmutation of minor actinides (with available fuels). During this period, MYRRHA will also be used as a flexible fast spectrum irradiation facility with the applications catalogue as described previously. It is also envisaged to deflect a small part (about 5% of the nominal beam) for nuclear physics applications at the proton accelerator.

Up until now, all flexible irradiation facilities have been operated very efficiently in critical mode. Working efficiently and reliably in subcritical mode introduces heavy constraints on the accelerator. Since working in critical mode is not conditioned by this fact, it might be advantageous to work in critical mode. The facility will then be converted into a critical fast spectrum irradiation facility based on heavy liquid metal coolant technology by separating the accelerator from the reactor, removing the spallation source and adding fuel assemblies to reach criticality. The entire accelerator facility can then be used for nuclear physics applications by the user-club of Belgian and other universities.

In the central positions, which host the spallation source, an in-pile section with a maximum high flux can be inserted. MYRRHA will then be operated as a flexible fast spectrum irradiation facility with the applications catalogue as described earlier, except for the demonstration of the ADS concept. Further efficient demonstration of minor actinides can be pursued in the critical MYRRHA and new minor actinide fuels can be tested and qualified.

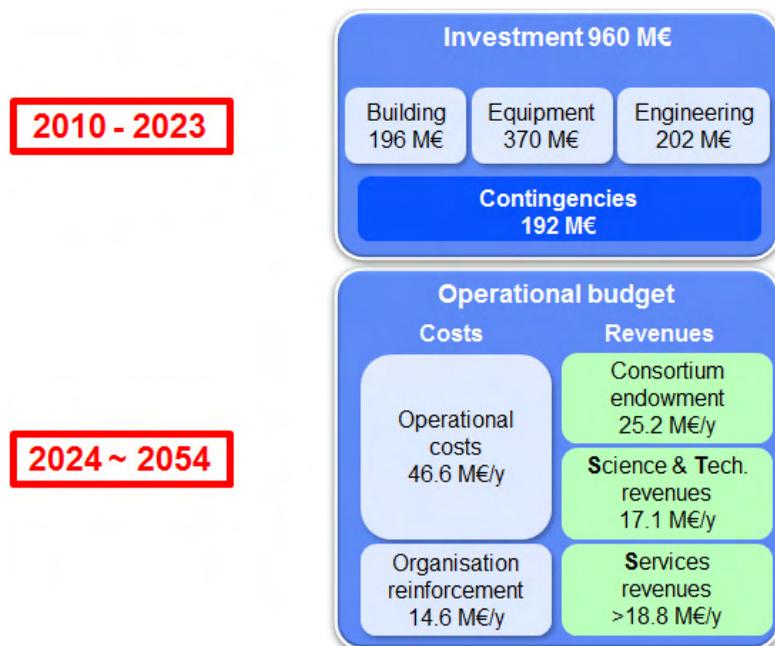
Presently nearly all major research facilities built worldwide are from the very beginning conceived as open-user facilities. Even the existing facilities that were built as national infrastructures are moving towards an open-user organisation. This means that the research facility proposed is of interest for the international research community and is unique or has characteristics making it very attractive to a large number of researchers and users. This leads to a formation of an owner consortium group that develops and later on operates the facility.

Apart from the traditional sources of funding, e.g. the contribution of the owner consortium group members during the investment phase of the project and the endowment for the operation phase, SCK•GEN management, as the promoter of the project, intends to call on the recent financing facility jointly developed by the European Commission (EC) and the European Investment Bank (EIB) in support of higher risk financing of research infrastructures such as the MYRRHA infrastructure.

The total investment costs (in EUR 2009) have been estimated [11] at EUR 960 M, including direct investment (civil engineering, equipments, components and instrumentation and control, spare parts), detailed design, installation co-ordination and licensing, project management costs, R&D support programme and about EUR 192 M of contingencies on the direct investment cost.

Similarly the operational costs (in EUR 2009) amount approximately to EUR 46.6 M including direct operational costs (labour for operations, fuel, waste management, provisions for dismantling, other operationally-linked services, contingency, financial costs) and the experiments' costs (labour for scientific experiments). A supplementary amount of about EUR 14.6 M should be foreseen for providing full support for technology and services development (reinforcing of internal organisation).

**Figure 5: Budget: balancing costs and revenues**



## Conclusion

Europe aims to become the most competitive and dynamic knowledge-driven economy, as decided in Lisbon in March 2000, by encouraging and promoting research and innovation. In addition, Europe is also facing the challenges of its long-term sustainable development by use of non-CO<sub>2</sub> emitting energy technologies that can play a significant role in guaranteeing the energy independence of Europe.

SCK•CEN is proposing to replace its ageing flagship, the Materials Testing Reactor BR2, by a new, flexible irradiation facility MYRRHA. In view of its innovative character and its field of application MYRRHA contributes to the realisation of the EU's goal towards a sustainable future.

MYRRHA is conceived as a flexible fast spectrum irradiation facility able to work in subcritical and critical mode. MYRRHA is foreseen to be in full operation by 2023 and it will be operated in the first years as an accelerator-driven system to demonstrate ADS technology and the efficient demonstration of minor actinides in subcritical mode.

Afterwards it is intended to decouple the accelerator from the reactor and run MYRRHA as a critical flexible fast spectrum irradiation facility by removing the spallation target. As a fast spectrum irradiation facility, it will address fuel research for innovative reactor systems, materials research for Generation IV systems and for fusion reactors, radioisotope production for medical and industrial applications, such as Si-doping. Being based on the heavy liquid metal technology, lead-bismuth eutectic, MYRRHA will be able to significantly contribute to the development of lead fast reactor technology. Since MYRRHA will also be operated in critical mode, it can even better play the role of European Technology Pilot Plant in the roadmap for LFR.

It is intended to organise the project as an international open-user facility to be constructed by an owner consortium made of European and even larger research centres and European member states' representative agencies or international bodies such as the European Commission.

### Acknowledgements

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## The Deep Burn project: 2010 overview and progress

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### Abstract

The Deep Burn (DB) project is a United States Department of Energy (DOE) feasibility study of transuranic management using high burn-up fuels in the high-temperature reactor (HTR). The project started in October 2008 and it is currently in its second year. The project is carried out by a team consisting of the Idaho National Laboratory (INL); Oak Ridge National Laboratory (ORNL); Argonne National Laboratory (ANL); Los Alamos National Laboratory (LANL); several universities, and other private entities.

The objective of the project is to establish the technology foundations needed to evaluate the role of the HTR in the nuclear fuel cycle, by providing the information needed to: evaluate cost-effective and economically attractive recycle options for light water reactor (LWR) fuel using the HTR high burn-up capabilities that will reduce transuranic (TRU) stockpiles; evaluate the capacity of geologic facilities to store HTR spent fuel, both low-enriched uranium (LEU) and DB fuel; consider the HTR as an important part of nuclear growth scenarios that include closing the fuel cycle; incorporate HTR fuel cycle issues into the mission of the next generation nuclear plant (NGNP).

Highlights of activities and achievements of the project will be presented, including:

1. *Core and fuel analysis.* The neutron energy spectrum in a DB-HTR differs from that in the ordinary HTR, which is based on the use of enriched uranium as the fissile material in the fuel. Therefore, modifications to the existing HTR core physics modelling tools had to be implemented. Analyses of both the prismatic and pebble-bed core types have been carried out that quantify the TRU destruction capability, the power and fuel temperature peaking in the core, and the temperature reactivity coefficients. The performance of the coated particle fuel is quantified in terms of the maximum stress in the silicon carbide coating layer and in terms of its potential consequent failure probability during irradiation in the core. The initial conceptual core designs have been optimised to retain acceptable predefined safety limits together with a high TRU destruction performance in excess of 60%.
2. *Spent fuel management.* The basic models required to determine radionuclide release from spent TRISO coated fuel were defined.
3. *Fuel cycle integration.* The DANESS code has been used to do preliminary modelling of the various ways that LWR, HTR and sodium fast reactors (SFR) can be combined to reduce TRU waste (synergy scenarios).
4. *Fuel modelling.* Recent advances in the understanding of materials behaviour and computing capability are used to enhance analytical modelling for high burn-up coated particle fuel. The task is divided into three subtasks: thermochemical modelling, transport phenomena and radiation damage.

5. *Fuel qualification. Progress has been made in laboratory-scale development of TRU TRISO-coated particle fuel, including kernel fabrication, fluidised bed coating, kernel and coated particle characterisation and analysis of ZrC material properties.*
6. *Spent fuel recycle. The recycle of HTR spent fuel is being investigated in the project to address the needs of LWR TRU recycle options under consideration (specifically the synergy scenarios), and to provide integration of the HTR into the fuel cycles investigated by the Fuel Cycle Research & Development (FCRD) programme.*

## Introduction

The HTR was one of six concepts selected in the 2002 *Technology Roadmap for Generation IV Nuclear Energy Systems* by the international community. In 2003, the DOE initiated development and demonstration of the HTR as a nuclear technology R&D priority, the NGNP, with hydrogen production identified early on in the roadmap as an important mission and the principal R&D focus. The NGNP programme goal is to construct an HTR prototype, with industry participation, by the year 2021.

The project mission for NGNP is to design, license and construct a commercial prototype gas-cooled reactor to demonstrate its capabilities. DOE, together with its lead nuclear energy laboratory (Idaho National Laboratory) and a coalition of end-users and vendors, is presently attempting to develop an industrial coalition to share in the cost of the prototype construction. The NGNP project includes a robust R&D programme (fuel, structural materials and methods), along with conceptual design studies in collaboration with industry. It is hoped that with the successful construction of the NGNP, large-scale deployment with applications in electricity production and process heat applications would result. Likewise, the reactor technology and infrastructure, designs and licensing, construction, and operating experience with the NGNP will provide the major part of the basis for designing, licensing and constructing DB modular helium reactors (MHR).

In parallel, the Generation IV International Forum (GIF) continues to conduct R&D for the longer term that would improve the economics and safety performance of subsequent commercial HTR. DOE is a major contributor to this HTR collaboration. In this context, with the challenges and lessons learnt from the existing LWR nuclear fleet, the issue of spent fuel has dominated much of the political and social discussion regarding nuclear power, and the lack of clarity and consensus regarding the disposition of spent fuel serves as a major obstacle in the expansion of nuclear power.

The Deep Burn project is an element of such an approach in which attention is devoted to the fuel cycle role of the HTR in terms of: i) using LWR spent fuel as feedstock for both HTR to reduce TRU in waste and minimise the necessary assured isolation interval; ii) recycle spent HTR fuel to further reduce TRU and the required isolation interval; iii) use HTR in conjunction with SFR to minimise transuranic deposition in a repository or any other means of deposition; iv) the long-term storage performance of spent HTR fuel in possible repository environments. As such, the project shares several of the objectives of the AFCI.

The near-term method of reducing the burden of spent LWR fuel at a lower cost, and the long-term benefits of reducing repository requirements and providing a means to reduce the ultimate size of the fuel cycle facilities are a major incentive to develop gas-cooled reactors with ultra-high burn-up coated particle (Deep Burn) fuel.

The examination of these aspects of the fuel cycle and resolution of any issues are essential to promoting a large-scale role for this reactor in the future deployment of nuclear power.

## The concept of Deep Burn

The term “Deep Burn” (DB) refers to the large fractional burn-up (up to 60-70% fissions per initial metal atoms [FIMA]) that can be achieved using transuranics with a single irradiation, in a

TRISO-fuelled gas-cooled reactor. The concept is particularly attractive because it would employ the same reactor design being considered for the NGNP, with the same potential for highly efficient electricity and hydrogen production, and other process heat applications. Spent TRISO fuel from DB can either be placed directly into long-term storage to provide containment of the residual radioactivity, or recycled in fast reactors. DB would rapidly and effectively reduce the inventory of TRU from spent fuel without the need for repeated recycles, precluding the possible weapons-related use of the residuals. In the past three years, with the help of high-fidelity neutronics models developed for the gas-cooled reactor, the basic physics of the DB concept have been thoroughly analysed and validated by universities, national laboratories and industry, both in the United States and worldwide. The Deep Burn project builds upon this work to systematically address how the HTR can leverage the features of its coated particle fuel to contribute to the minimisation of TRU waste.

### **The importance of TRISO fuel**

The key to the HTR behaviour and performance with respect to TRU management is its TRISO fuel. This engineered fuel form (microfuel) is made of small particles having a 200-400 micron fuel core surrounded by carbon and silicon carbide structural layers that can contain the fuel and fission products under all normal and off-normal reactor events, thus giving rise to the “inherently safe” character of the reactor.

During DB irradiation, the various constituents of the TRU TRISO fuel particle are progressively destroyed according to their cross-section. For instance, at the 50% burn level, almost all the  $^{239}\text{Pu}$  (90%) has already been destroyed and, thus, the risk of diversion of spent fuel for weapons production is also eliminated. Of crucial importance to the DB concept is the interplay between  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Pu}$ .  $^{239}\text{Pu}$  is a strong fissile isotope, while  $^{240}\text{Pu}$  is a strong burnable poison that is also a fertile isotope producing  $^{241}\text{Pu}$ . The utilisation of the short-lived  $^{241}\text{Pu}$  in the high-burn-up TRISO fuel without reprocessing is a special feature of DB. The sequence 239-240-241 produces a very steady reactivity behaviour over long periods of time leading ultimately to the achievement of better than 60% FIMA burn-ups without exceeding TRISO fuel fast neutron fluence limits.

Although the DB TRISO fuel shares common elements with the TRISO fuel proposed for the NGNP TRISO fuel and the joint Russian National Nuclear Security Administration (NNSA) excess weapons Plutonium Disposition Programme (Pu-TRISO fuel), manufacturing of TRU TRISO will require the additional implementation of remote handling techniques and the development of fuel designs specifically geared to very high burn-up.

### **Value of DB in the fuel cycle**

In parallel with the physics analysis, preliminary work has indicated that because of the large amount of useful energy that can be extracted from the DB TRISO fuel (up to 20 times greater than from mixed-oxide [MOX] fuel in LWR), it may be possible to recover all or part of the costs of reprocessing LWR spent fuel. The DB concept potentially creates a different paradigm for the near-term economics of closed fuel cycles, if the cost of spent LWR fuel reprocessing can be offset by the value added through the recovered TRU, producing power at a competitive cost.

### **Spent fuel recycle and disposal**

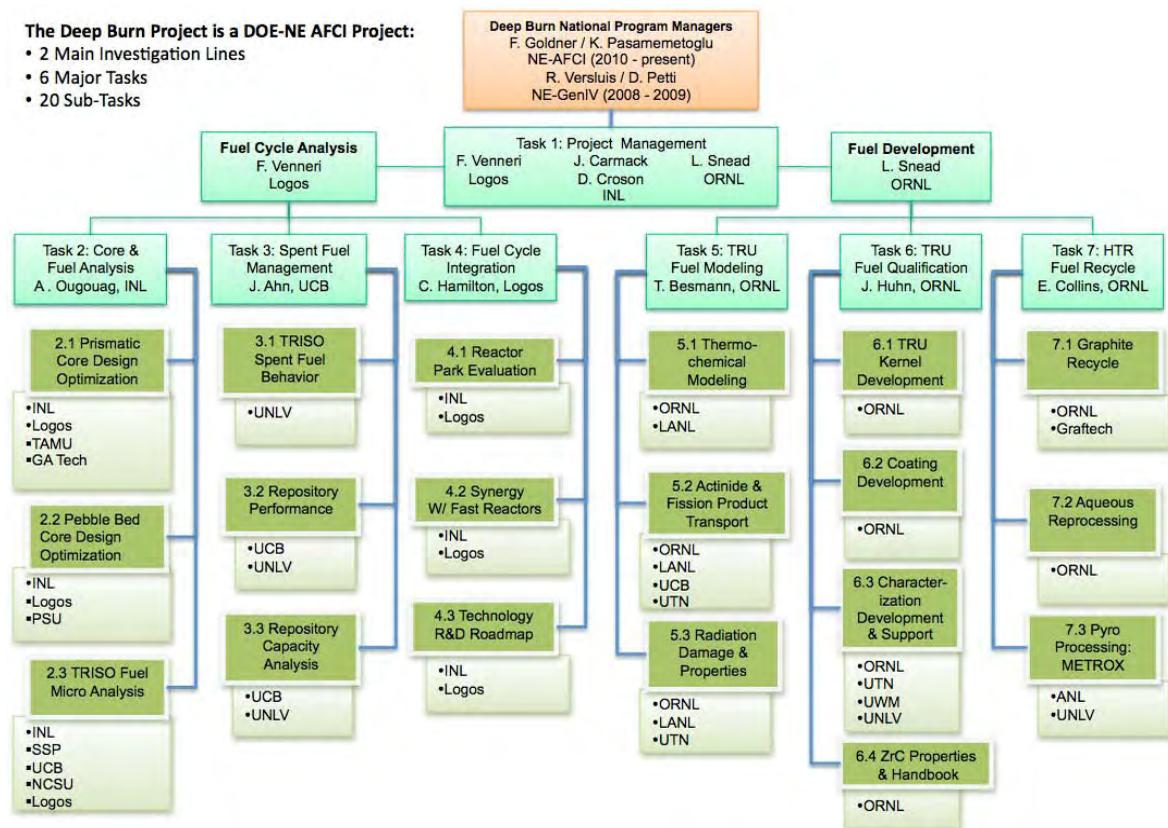
Spent TRISO fuel, be it from HTR LEU operations or DB LWR-TRU fuels, can be directly disposed of in a repository or further recycled in fast reactors. For the long-term disposal option, a clear understanding of the performance of TRISO fuel in the repository is required, so that a repository strategy for TRISO fuel can be developed, taking into account the possibility of larger volumes, but also the capability to much better retain the radioactive isotopes afforded by the ceramic fuel. In the recycle option, the addition of a TRISO “head-end,” such as “grind and leach” or metal

recovery from oxide fuel (METROX), to the mainstream partitioning schemes (UREX+ or PYRO) would be required, but the relevant technologies need to be developed. In addition to fuel recycle options, the recycle of spent fuel graphite offers the potential for a small waste stream from the HTR.

### Project structure

Project work is divided into six technical tasks organised under the work breakdown structure (WBS) presented in Figure 1, which also identifies the national laboratories, universities and private sector organisations working on the various tasks.

Figure 1: DOE Deep Burn project



### Core and fuel analysis

The initial conceptual reference designs have been completed for both the pebble bed and prismatic DB-HTR variants. The TRU destruction performance was found to be as expected, with average burn-ups in excess of 60%. During the preliminary reactivity and safety analysis, several new findings were made, among them:

- The increased decay heat from the TRU in the fuel appears to be an important constraint in the design of DB-HTR so that excessive temperatures are not reached in depressurised conditions.
- TRU-fuelled HTR cores exhibit higher power peaks, compared to the low-enriched uranium (LEU)-fuelled core versions requiring the use of additional burnable poisons.

A first iteration on the initial reference designs has provided the means to address these and other issues raised in the analysis of the initial references.

### *Spent fuel management*

The basic models required to determine radionuclide release from spent TRISO-coated fuel were defined by the University of California at Berkeley (UCB) and the University of Nevada-Las Vegas (UNLV). These models included algorithms for calculating:

- Coating failures of spent particles, as a function of time, caused by corrosion and/or stresses from internal gaseous fission products and helium pressure.
- Dissolution-rate controlled release from the fuel kernel.
- Reactive transport through the sorptive graphite matrix. Preliminary measurements of the sorption coefficients of graphite required for the models have been completed by UNLV.

The initial results from these evolving models suggest that coated particle spent fuel is potentially a significantly superior waste form, but that the several rate-dependent mechanisms noted in the previous description of the models must be quantitatively evaluated to ensure that the advantages are confirmed. Preliminary calculations comparing the radiotoxicity of the total inventory of canisterised spent nuclear fuel (CSNF) to spent once-through DB fuel indicates that the reduced TRU content in the spent DB fuel results in a reduction in toxicity per GWe-year produced, due to the higher efficiency of the HTR. More importantly, when the retention capability of the DB spent fuel is included, the radiotoxicity of material released from the spent DB HTR fuel is ~100 times less than the CSNF material. Graphite appears to play a role comparable to that of the TRISO fuel in holding up fission products in the repository.

### *Fuel cycle integration*

The project modelled the various ways that LWR, HTR and sodium fast reactors (SFR) can be combined to reduce and potentially minimise TRU waste. For the simplest application of DB in HTR (LWR spent fuel TRU inventory being used to fuel a DB-HTR), spent LWR TRU is reduced by 70% and weapons-useful plutonium is effectively eliminated. When SFR are subsequently used to continue to irradiate the residual TRU from DB-HTR (synergy scenario), only trace amounts of TRU need to be sequestered in a repository. In such a scenario, the HTR reduces, by 60% to 90%, the amount of spent fuel recycling required to support the primary power producing LWR. Most importantly, DB scenarios (including the synergy scenario) involve substantially smaller inventories of TRU in the fuel cycle, fewer recycling facilities, and are at least twice as effective as other scenarios at actually destroying TRU.

The preliminary technology research and development (R&D) roadmap for the DB-HTR has been developed in co-ordination with all members of the project team, but also in co-ordination with the NGNP, Global Nuclear Energy Partnership (GNEP) and Advanced Fuel Cycle Initiative (AFCI) programmes. This co-ordination allows the project to avoid duplication of technology activities being addressed in these related programmes and focus their technology roadmap activities on the incremental development required for the DB-HTR. The activities include establishing a Technology Readiness Level (TRL) scale, assigning a preliminary TRL for critical technologies and determining the general design data needs (DDN) in each area.

### *Fuel modelling*

ORNL has co-ordinated the development of the research plan for exploiting recent advances in the understanding of materials behaviour and computing capability to enhance analytical modelling for high-burn-up coated particle fuel. The task is divided into three subtasks: thermochemical modelling, transport phenomena and radiation damage.

The thermochemical evolution of a DB tri-structural isotropic (TRISO) fuel particle designed for the consumption of plutonia and other transuranics was modelled. The results show that oxygen-to-metal (O/M), and therefore oxygen pressure/potential, increases with burn-up because of released oxygen that is not fully taken up by forming fission product oxides. The presence of gettering phases such as SiC (silicon carbide) or ZrC (zirconium carbide) greatly reduces the O/M and oxygen pressure as well as CO (carbon monoxide) and CO<sub>2</sub> (carbon dioxide) pressures.

Gettered kernels were identified as the best way to control the oxygen potential in the coated particle and offer the best path to high burn-up.

The formation energies of a range of point defects and palladium-defect complexes in SiC have been calculated from first principles. The complete set of ZrC point defect structures and formation energies were calculated for the first time by means of *ab initio* calculations.

#### *Fuel qualification*

Progress has been made in ORNL's laboratory-scale development of TRU TRISO-coated particle fuel, including kernel fabrication, fluidised bed coating, kernel and coated particle characterisation, and analysis of ZrC material properties; the use of sol-gel processes for fabricating TRU kernels has been assessed; conceptual design has been completed for a fluidised bed coater for TRU TRISO using glove box containment; tests were performed on three different Zr delivery systems for the Zr coater; measurement of silver diffusion, palladium attack and high-temperature material properties has been initiated at the responsible institutions, University of Wisconsin (UW), UNLV and ORNL. From the work performed this year, it appears that ZrC is hard to fabricate via CVS, hard to characterise in coated particles, and hard to experiment with, since slight deviations in stoichiometry and oxygen pick-up in the surface of the carbide can make experiments difficult to carry out and interpret. Furthermore some of its high-temperature properties appear problematic, and in the final analysis, may not be needed for DB fuel.

#### *Spent fuel recycle*

The recycle of HTR spent fuel is being investigated in the project both to address the needs of LWR TRU DB recycle options under consideration (specifically the synergy scenario), and more generally to provide integration of the HTR into the fuel cycles investigated by the AFCI. Dry and wet separation processes are being considered.

Independent of the downstream separations process chosen, graphite recycle is important to reduce the waste stream from such operations. The graphite recycling effort was started with the formulation of a plan of action, the completion of an inventory of the available irradiated graphite at ORNL, and the start of the required operations on non-irradiated graphite. Based on initial activities, the recycle of irradiated spent fuel graphite appears viable.

## Systematic evaluation of uranium utilisation in nuclear systems

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### Abstract

*The uranium utilisation values of fast-spectrum once-through breed-and-burn core concepts have been evaluated in this study. Examples of these breed-and-burn systems include the CANDLE concept, the travelling wave reactor (TWR) concept of TerraPower LLC, the ultra-long life fast reactor (ULFR), and the sustainable sodium-cooled fast reactor (SSFR). In order to provide a systematic understanding of these systems, scoping evaluations are first performed and the uranium utilisation values are calculated and compared with that of the once-through PWR system. For the PWR system, the uranium utilisation is less than 1% regardless of the burn-up because a large fraction of the source natural uranium becomes depleted uranium during the enrichment process, rather than used for energy generation by fission. With a favourable sustainable capability with fast neutrons, however, the breed-and-burn core concepts can use the uranium resource effectively for generating energy. The attainable uranium utilisation of the once-through breed-and-burn core concepts is as high as ~30%, though some technical design issues, such as long transient period, high neutron fluence, big core size, etc., must be resolved in order for these core concepts to be practical.*

## Introduction

In the United States, sustainable fuel cycle options are being developed for improving uranium resource utilisation and reducing the proliferation risk and the used nuclear fuel waste at the same time [1]. The uranium utilisation is defined as the ratio of the heavy metal mass burnt by fission to the total uranium mass used for making the fuel. High uranium resource utilisation is expected to support national energy security and climate change goals by effectively using uranium resources for nuclear energy generation.

Uranium resource utilisation is dependent on the design choices of the nuclear system and the applied fuel cycle strategies. For the full-recycling (i.e. fully-closed) fuel cycle option with a breeder reactor, the natural uranium resource is required only for making the enriched uranium fuel of the initial core, and the subsequent passes could be sustainable by recycling the used nuclear fuel. Thus, the uranium utilisation could theoretically be more than 95% under the full recycling fuel cycle option [2]. Due to issues that have been attributed to the full recycling fuel cycle options, however, nuclear energy is mostly generated by once-through thermal systems (LWR). These issues include proliferation concerns, reactor design and fuel cycle costs.

The modified open fuel cycle option is also being explored in the United States. This is any fuel cycle option lying between the extremes of the once-through fuel cycle option and the full recycling fuel cycle option. It is expected that the uranium utilisation of the modified open cycle nuclear systems is higher than that of the once-through nuclear systems, but smaller than that of the full recycling nuclear systems. No evaluation of the uranium utilisation of the modified open cycle was performed in this study.

Compared to the modified open or full recycling fuel cycle options, the uranium utilisation of the existing commercial once-through nuclear systems such as pressurised water reactor (PWR) is very small because most of the source natural uranium becomes depleted uranium during the enrichment process, rather than used for energy generation by fission. As a result, the existing commercial once-through nuclear systems use less than 1% of the energy available in the mined uranium [2].

To improve the uranium utilisation under the once-through fuel cycle option, one approach being considered by nuclear reactor designers is the use of a breed-and-burn concept based on fast reactor technologies. With a favourable breeding capability with fast neutrons, the breed-and-burn concept targets a sustainable fuel cycle using depleted uranium, in which the enriched uranium or other fissile material is required for the initial core operation, and subsequent passes require only depleted uranium for maintaining criticality.

In recent years, advanced breed-and-burn core concepts have been proposed for the effective utilisation of uranium resources. Examples of systems include the CANDLES concept [3] by the Tokyo Institute of Technology, the travelling wave reactor (TWR) concept of TerraPower LLC [4], the ultra-long life fast reactor (ULFR) by ANL [5], the fast mixed spectrum reactor (FMSR) by BNL [6], and the sustainable sodium-cooled fast reactor (SSFR) by ANL. Theoretically, the uranium utilisation of these core concepts could be increased to their average discharge burn-up at the equilibrium state because the external natural uranium resource is only required for the initial core and the subsequent cycles can be sustainable using depleted uranium.

The breed-and-burn characteristics and attainable uranium utilisation are dependent on the design choices. The CANDLES and ULFR core concepts employ a one-batch fuel management scheme: the whole-core nuclear fuels would be charged and discharged at the beginning of reactor life (BOL) and the end of reactor life (EOL), respectively. These systems have tended to propagate the burn zone from the ignition region to a blanket region. The ignition (or starter) region contains driver fuel that has fissile material, while the blanket region contains fertile material that is depleted uranium. The ignition region is designed to allow the breeding of sufficient fissile material in the adjoining blanket region. As the irradiation time progresses, the power in the ignition region decreases as the fissile material burns and fission products are accumulated, and the power in the adjoining blanket region increases due to the bred plutonium. As a result, the burn zone moves to the blanket region from the initial ignition region.

The SSFR and TWR core concepts adopt a multi-batch fuel management scheme, in which the burnt fuels are partially replaced with fresh depleted uranium fuels. The number of batches and cycle length were determined such that the core can maintain criticality by feeding depleted uranium fuel only at the equilibrium state: generally, the number of batches is higher than 30 and the fuel residence time is longer than 50 years.

In order to provide a systematic understanding, scoping evaluations have been performed and the uranium utilisation values of these advanced once-through nuclear systems have been compared to those of existing once-through thermal reactors. It is noted that the scoping calculations were performed using open literature data or reasonable guesses. Thus, some results of this study may differ from the values generated by the concept developers; furthermore, some of the concepts are still evolving.

## Advanced breed-and-burn core concepts

### *CANDLE core concept*

The “Constant Axial Shape of Neutron Flux, Nuclide Density and Power Shape During Life of Energy Production” (CANDLE) reactor concept has been considered for very high uranium fuel utilisation. The concept was proposed by researchers at the Tokyo Institute of Technology [3]. The reactor design has an ignition region at the core bottom and a tall axial depletion region above the ignition region. The ignition region (starter) is used for initial power generation and for the ignition of power generation in the depletion region. This is accomplished by the use of leaking neutrons to breed fissile material in the depletion region.

The scoping calculation of the CANDLE core concept was performed using cylindrical core geometry. The height of the starter is 120 cm and it is designed to have different low-enriched uranium (LEU) fuels axially to enhance the axial propagation of the burn zone: 13%, 7% and 3% from the core bottom with lengths of 80 cm, 20 cm and 20 cm, respectively. A depletion region of height 6.8 m, which contains depleted uranium, is located above the starter. The core has a diameter of 4.0 m and is surrounded by a 50 cm thick radial reflector made of depleted uranium. For the sake of simplicity, it was assumed that all assemblies have the same fuel, coolant and structure volume fractions of 37.5%, 30.0% and 20%, respectively.

The CANDLE reactor has a power rating of 3 000 MWt with sodium coolant and U-Zr fuel. The core performance parameters are provided in Table 1. Initially, the core multiplication factor increases slightly and decreases as  $^{235}\text{U}$  is burnt, but it stabilises with excess reactivity of  $\sim 1\% \Delta k$  as  $^{239}\text{Pu}$  is bred in the depletion region. Most of the core power is initially generated in the starter region. With the increase in the irradiation time, the power density of the starter region decreases due to the poisoning effect of fission product accumulation. Consequently, the burn zone moves to the neighbouring depletion region. This burn zone propagates to the top of the core with a moving speed of  $\sim 3.5$  cm per year. Thus, the core can maintain criticality for  $\sim 200$  years. The average burn-up of the depletion (starter and axial blanket) region is about 40%, and the overall average burn-up including the radial blanket is about 25%.

**Table 1: Core performance parameters of CANDLE**

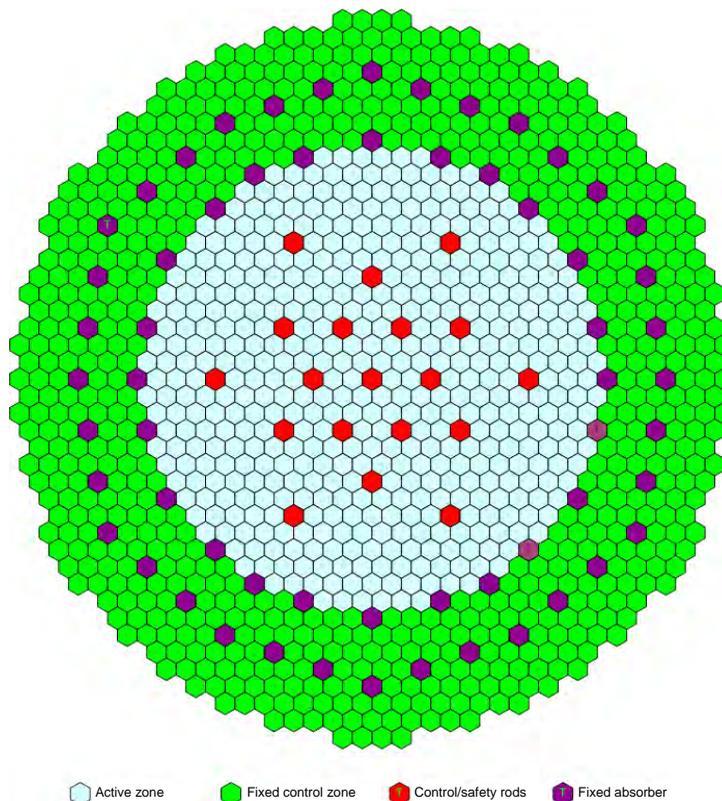
Parameter	Value
Thermal power, MWt	3 000
Core height including depletion region, m	8.0
Attainable cycle length with full power, year	200
Average enrichment of starter fuel, %	10.3
Specific power density, MW/t	3.7
HM inventory of total core, tonne	820
Average burn-up (starter/depletion/reflector), GWd/t	362/396/25
Peak discharge fast fluence, $10^{23}$ neutrons/cm <sup>2</sup>	41.9

### Travelling wave reactor concept

The travelling-wave reactor (TWR) is an advanced concept proposed for extending uranium resources. It employs a core containing a nuclear ignition region and a burn-wave propagation region containing natural uranium. It has been proposed that the core be placed ~100 meters underground [7]. However, the TWR proposed by TerraPower LLC is a special class of travelling wave core design, i.e. a *standing* wave of breeding and burning zones is established by periodically moving core material in and out of the breed-burn region instead of the wave moving through a fixed core material.

Figure 1 shows a conceptual drawing of the TWR core concept at the beginning of life (BOL). The core is approximately a cylindrical core geometry composed of hexagonally-shaped fuel bundles, or assemblies, containing a combination of enriched and depleted uranium fuel pins clad in ferritic-martensitic steel tubes. The core adopted a metallic fuel with 5-8% zirconium to dimensionally stabilise the alloy during irradiation and to inhibit low-temperature eutectic and corrosion damage of the cladding. The fissile assemblies are primarily located in the central core region, designated the Active Control Zone (ACZ), which generates most of the core power. Fertile assemblies are primarily placed in the core peripheral region, called the fixed control zone (FCZ) and their number is selected such that reactor operation is possible for about 40 years without the need to bring new fuel into the reactor from the external feed. In addition, the FCZ also contains a sufficient number of spare fissile and fertile fuel assemblies in case replacement assemblies are needed for failed fuel pins. The initial core loading is configured to ensure criticality with a small amount of excess reactivity and ascension to full power output shortly after initial reactor start-up. Excess reactivity monotonically increases because of breeding until a pre-determined burn-up is achieved in a selected number of fuel assemblies. The reactivity increase is compensated by control rods, which are gradually inserted into the core to maintain core criticality.

Figure 1: Conceptual radial layout of TWR at beginning of life



After a pre-determined amount of time, the TWR is shut down in order to move high-burn-up assemblies to the FCZ near the core periphery replacing them with depleted uranium assemblies. This fuel shuffling operation is expected to take one to two weeks depending on the number of fuel assemblies requiring shuffling. Over the reactor life, it might be necessary to move the spent fuel assemblies more than once to achieve adequate breeding ratio and to minimise damage to the fuel assemblies. Based on the proposed fuel management schemes, the core performance parameters are presented in Table 2. In this calculation, fuel re-purposing (partial or mechanical separations) was not considered and the core was shuffled 15 times with an assumed cycle length of 800 days.

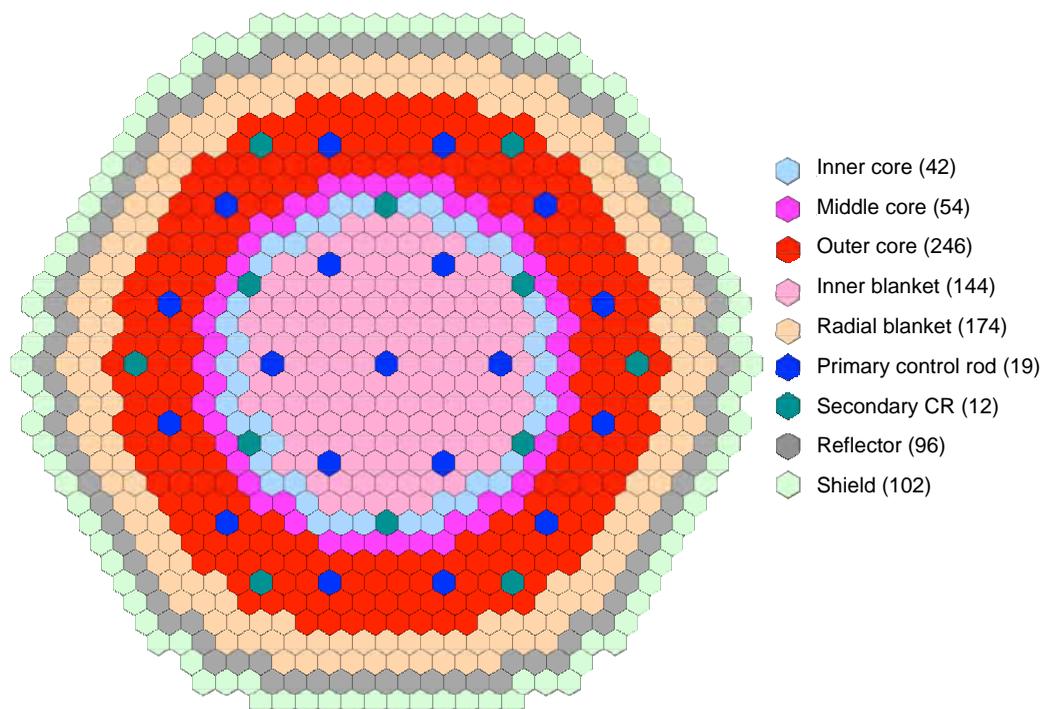
**Table 2: Core performance parameter of TWR**

Parameter	Value
Thermal power, MWt	3 000
Core height including axial blanket, cm	250
Specific power density, MW/t	7.5
Average discharge burn-up, including FCZ fuels, %	9.8

### **Ultra-long life fast reactor core concept**

The ultra-long life sodium-cooled fast reactor (ULFR) concept was developed aiming for a reactor operation without refuelling over a long reactor lifetime [6]. The power density is significantly de-rated to increase the reactor operation time without refuelling. Figure 2 shows the radial core layout of ULFR, which is a unique annular core layout. All internal blanket assemblies are located at the core centre, and are surrounded sequentially by driver assemblies and radial blankets. Based on this core configuration, the power is generated in the driver region when the reactor irradiation starts, but the burn zone moves inwardly from the exterior driver region into the interior blanket region as the irradiation progresses.

**Figure 2: Radial layout of ULFR**



For the ULFR, molybdenum-based alloy (U-<sup>10</sup>Mo) was selected in order to increase the heavy metal loading in the core. In order to achieve inward power (burn-up) propagation, different uranium enrichments have been used for the driver fuels, varied along the core radial direction. The enrichment of the inner, middle, and outer core zones is 9%, 11% and 13%, respectively. Depleted uranium fuel with <sup>235</sup>U content of 0.25% is loaded into the internal, axial and radial blanket core regions. The assembly design parameters were determined such that the reactor can maintain criticality for more than 50 years without refuelling and the peak excess reactivity can be controlled by a limited number of control assemblies.

The core performance parameters for the ULFR are provided in Table 3. The ULFR core can maintain criticality for 54 years with a capacity factor of 90%. The average discharge burn-up of the driver fuel is 316 GWd/t with the peak fast fluence of  $\sim 22 \times 10^{23}$  n/cm<sup>2</sup>, while that of the blanket fuel is 95 GWd/t. The power sharing of the driver fuel is 94% at BOL, but decreases to 27% at the end of life, due to the movement of the primary burn zone during the reactor operation.

**Table 3: Core performance parameters of ULFR-3000**

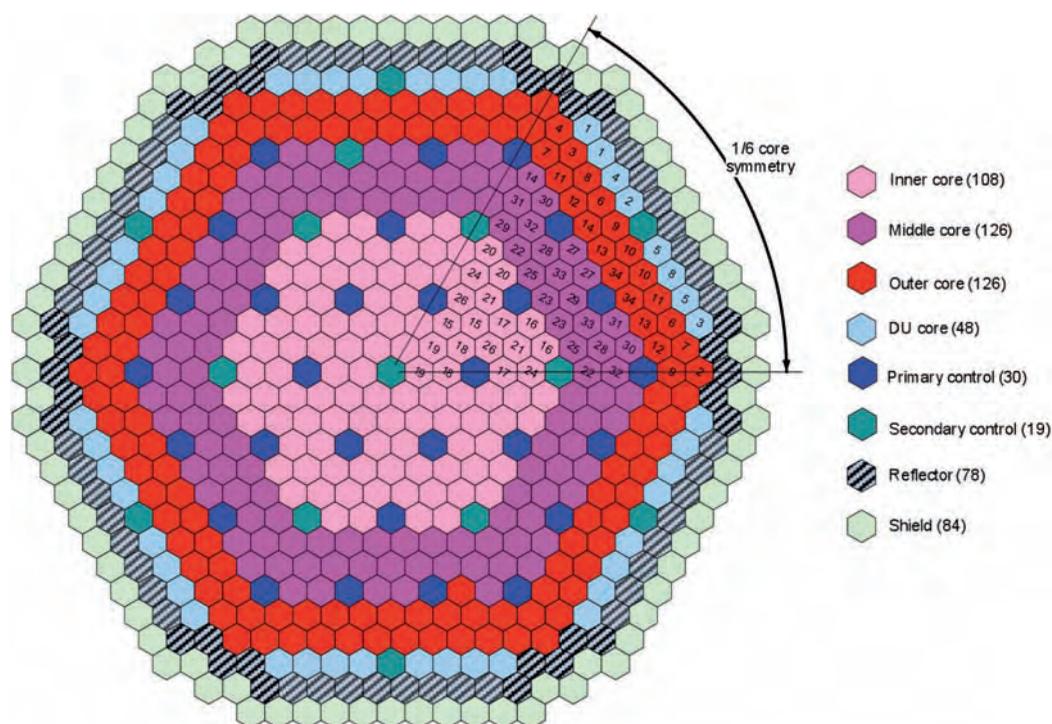
Parameter	Value
Thermal power, MWt	3 000
Cycle length without refuelling, year	54
Initial heavy metal inventory (driver/blanket), tonne	103/217
Discharge burn-up (driver/blanket), GWd/t	316/95
Peak excess reactivity, %Δk	3.87
Peak discharge fast fluence, 10 <sup>23</sup> neutrons/cm <sup>2</sup>	22.1
Average power density (driver/blanket), W/cm <sup>3</sup>	109.1/33.1
Power sharing of driver fuels (BOL/EOL), %	94/27

### **Sustainable sodium-cooled fast reactor**

A sustainable sodium-cooled fast reactor (SSFR) core concept was developed targeting a sustainable fuel cycle with depleted uranium feed only. The core design parameters and fuel management scheme were determined such that the depleted uranium should be irradiated for a certain period until sufficient plutonium is bred. The resulting fuel management scheme of the SSFR core concept is a 34-batch scheme with the cycle length of 1.5 years and the capacity factor of 90%. Figure 3 shows the radial core layout, which consists of 408 driver fuels. Thus, twelve burnt assemblies are replaced with the fresh fuel (which is the depleted uranium fuel) every 1.5 years and the fuel resides in the core for 51 years. The sequential fuel movements based on the 60° symmetric core are indicated in Figure 3.

The core performance parameters are provided in Table 4. The core multiplication factor is stabilised after 60 cycles with excess reactivity of 3.1%Δk at the equilibrium cycle. As indicated in Figure 3, the depleted uranium fuel is charged into the core periphery where the power density is relatively small and moves into the central active zones. Thus, the burn-up is small for the first 14 cycles. However, as the fuel is shuffled into the active core zones, the burn-up increases and becomes 280 GWd/t (peak fast fluence of  $26.7 \times 10^{23}$  n/cm<sup>2</sup>) when the fuel is discharged after 34 cycles.

It is noted that a fast mixed spectrum reactor (FMSR) was proposed by BNL in the 1970s targeting a sustainable operation with depleted uranium feed only [6]. The idea of the FMSR is to charge depleted uranium into the thermal core zone for breeding plutonium and shuffle it into the fast core zone as sufficient plutonium is bred. The feasibility of the FMSR core concept was also evaluated in this study, but the detailed core performance parameters are not provided here because it was observed that the core performance characteristics are generally comparable to those of the SSFR core concept.

**Figure 3: Radial core layout of SSFR**

**Table 4: Core performance parameter of SSFR**

Parameter	Value
Thermal power, MWt	3 000
Cycle length, year	1.5
Number of batches	34
Initial heavy metal inventory, tonne	177.6
Discharge burn-up, GWd/t	276.6
Peak discharge fast fluence, $10^{23}$ neutrons/cm <sup>2</sup>	26.7
Average power density, W/cm <sup>3</sup>	96.2

### Uranium utilisation of breed-and-burn core concepts

The uranium utilisation of the breed-and-burn core concepts were provided in Table 5, including the PWR with the discharge burn-up of 50 GWd/t (PWR-50) as the representative of the commercial once-through thermal reactors. In this study, the uranium utilisation is defined by the ratio of the heavy metal mass (HM) burnt by fission to the total uranium mass used for making the fuel.

For consistent comparison, the HM mass flow was normalised per year although whole-core fuel would be charged and discharged at the BOL and EOL in the one-batch fuel management scheme, which was adopted by the CANDLE, ULFR and TWR concepts. It is noted that the TWR is considered as a one-batch system in Table 5 although fuel shuffling occurs 15 times; the fuel shuffling is between the active core zone and fixed core zone, and the used fuel discharge occurs at the end of reactor life.

In Table 5, the HM burnt mass by fission is comparable (which is about one metric tonne per year) because the power ratings are all the same (3 000 MWth), but the required HM mass is dependent on the design choices. The required HM mass for the commercial PWR systems is

**Table 5: Comparison of uranium utilisation values**

	PWR-50	CANDLE	SSFR	ULFR	TWR
Reactor power, MW-thermal	3 000	3 000	3 000	3 000	3 000
Reactor power, MW-electric	1 000	1 200	1 200	1 200	1 135
Thermal efficiency, %	33.3	40.0	40.0	40.0	37.8
Reactor capacity factor, %	90	90	90	90	93
Neutron spectrum	Thermal	Fast	Fast	Fast	Fast
Fuel form	UOX	U-Zr	U-Zr	U-Mo	U-Zr
Tail uranium enrichment, %	0.25	0.25	0.25	0.25	0.30
Number of batches	3	1	34	1	1
Average burn-up, GWd/t	50	258	277	166	93
Cycle length per batch, year	1.5	200.0	1.5	54.0	38.0
HM inventory, tonne	89.0	823.7	177.6	319.6	399.3
HM charge (LEU/DU), t/yr	19.8/–	0.4/3.7	–/3.5	1.9/4.0	1.7/8.8
U enrichment (LEU/DU), %	4.21/0.25	10.3/0.25	–/0.25	12.2/0.25	14.0/0.3
Requires HM, t/yr	166.1	8.5	3.5	48.3	55.5
HM burnt by fission, t/yr	1.03	1.03	1.02	1.04	1.03
Uranium utilisation, %	0.6	12 <sup>a</sup>	29 <sup>b</sup>	2.2 <sup>a</sup>	1.9

<sup>a</sup> Uranium utilisation of the particular core concepts that are introduced in this paper. The maximum attainable uranium utilisation could be increased to the fuel discharge burn-up when the core size is increased.

<sup>b</sup> Uranium utilisation decreases when the transition cycle is accounted for in the calculation methodology.

dependent on the discharge burn-up. To achieve the discharge burn-up of 50 GWd/t, the PWR system requires 19.8 tonnes of 4.21% enriched uranium every year, which requires 166.1 tonnes of natural uranium. Thus, the PWR-50 system uses only 0.6% of natural uranium resource to generate energy. The discharge burn-up of the PWR system does not affect the uranium utilisation because more enriched uranium is required to increase the discharge burn-up. As a result, the uranium utilisation of the existing commercial once-through thermal systems is generally less than 1.0%.

For the CANDLE concept considered in this study, the core requires 0.4 tonnes of 10% enriched uranium for the starter fuel, which implies 8.5 tonnes of natural uranium assuming 0.25% <sup>235</sup>U in depleted uranium. Thus, the uranium utilisation of the CANDLE core concept is ~12%. In principle, the CANDLE concept can burn all depleted uranium resulting from the creation of the initial enriched uranium fuel of the starter zone by increasing the core height because the core can maintain criticality as long as depleted uranium is available. Using all depleted uranium arising from making the starter fuel, the maximum core height would be 15.3 m. For this case, the reactor can maintain criticality for about 430 years with a core power rating of 3 000 MWt and the attainable uranium utilisation is ~28%.

At the equilibrium state, the SSFR core can be sustainable without enriched uranium. The core requires 3.5 tonnes of depleted uranium and burns 1.04 tonnes of heavy metal. As a result, the uranium utilisation is about 29% at the equilibrium state. However, the uranium utilisation decreases if the transient cycle is counted. Thus, in order to achieve the maximum attainable uranium utilisation, additional activities on design fixes are required.

The ULFR core concept requires 1.9 tonnes of 12.2% enriched uranium for the driver fuels, which requires 48.3 tonnes of natural uranium. Thus, the uranium utilisation of the ULFR core concept is about 2.2%. It is noted that the uranium utilisation and the discharge burn-up can be increased by increasing the core diameter; however, further increase of the core diameter was not attempted because it would have a negative impact on the capital cost.

The TWR core concept requires 1.7 tonnes of 14% enriched uranium for the active control zone fuels, which requires 55.5 tonnes of natural uranium with the assumed tail uranium enrichment of 0.3%. Thus, the overall uranium utilisation is ~1.9%. TerraPower LLC is considering

the “re-purposing” (or re-cladding or re-conditioning) of the fuel following use. This is to allow the fuel burn-up in a given pass through the core to be increased to a much higher value (about 50%). This re-purposing could involve a simplified reprocessing step. In the paper by Ellis *et al.*, the proposed approach is melt refining [4]. This is a new direction for the TWR concept. Based on this approach, TerraPower LLC has indicated ~40-fold improvement in attainable fuel utilisation (~24% uranium utilisation) with a well designed TWR using a melt-refining process.

## Conclusions

There has been recent interest in approaches for increasing the uranium utilisation, while at the same time reducing the proliferation risk associated with the use of nuclear power. One approach being considered by reactor designers is the use of breed-and-burn concepts in which fissile material is produced in the fertile fuel region for the purpose of prolonging the irradiation time and increasing the burn-up during residence in the reactor. Most of the concepts involve the propagation of the active burn zone in the reactor core, either axially or radially. Some involve delicate assembly shuffling to enhance the fissile material breeding and to utilise the bred material efficiently. Examples of these breed-and-burn systems include the CANDLE concept by the Tokyo Institute of Technology, the travelling wave reactor (TWR) concept of TerraPower LLC, the ultra-long life fast reactor (ULFR) by ANL, and the sustainable sodium-cooled fast reactor (SSFR) by ANL. In order to provide a systematic understanding of these core concepts, the uranium utilisation values have been evaluated by using the core performance parameters obtained from scoping calculations.

The CANDLE and ULFR core concepts employ a one-batch fuel management scheme: the whole-core fuels would be charged and discharged at BOL and EOL, respectively. These systems have tended to propagate the burn zone from the ignition region to a blanket region. For the CANDLE reactor, the ignition region is isolated at the core bottom, and the blanket region is located above the ignition region. As a result, the burn zone moves axially along the axial blanket region. The ULFR core adopted an annular core concept, in which the central blanket region is surrounded by the driver region. Thus, the burn-zone is propagated radially. For the particular core concepts introduced in this paper, the CANDLE core concept uses about 12% of natural uranium to generate energy, while the ULFR uses 2.2%. However, since the uranium utilisation values of the CANDLE and ULFR concepts are dependent on the core size (in particular, depleted uranium inventory in the blanket region), the uranium utilisation could be increased to ~30% by increasing the core size.

The TWR proposed by TerraPower LLC is a special class of travelling wave reactor design; a standing wave of breeding and burning zones is established by periodically moving core material in and out of the breed-burn region instead of a wave moving through a fixed core material. The uranium utilisation of the currently proposed TWR core concept is ~1.9%; however, TerraPower LLC has indicated the possibility of achieving ~40-fold improvement in fuel utilisation (compared to the commercial PWR) by re-cladding the used nuclear fuel using the melt-refining process.

The SSFR core concept adopts a multi-batch fuel management scheme, in which the burnt fuels are partially replaced with fresh depleted uranium fuels. The number of batches and cycle length were determined such that the core can maintain criticality by feeding depleted uranium fuel only at the equilibrium state. At the equilibrium state, the uranium utilisation of this concept can be increased to its average discharge burn-up (29%) because the external natural uranium resource is required only for the initial core and the subsequent passes can be sustainable using depleted uranium. However, it is emphasised that it takes a long time to achieve such high uranium utilisation because the system slowly converges to the equilibrium cycle.

In conclusion, the uranium utilisation of the commercial once-through thermal system is less than 1% regardless of the discharge burn-up, but could be increased to ~30% in the once-through breed-and-burn core concepts. There are, however, technical design issues (such as long transient period, high neutron fluence, big core size, etc.) that would have to be resolved in order for these core concepts to be practical.

### Acknowledgements

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