Actinide and Fission Product Partitioning and Transmutation

12th Information Exchange Meeting
Prague, Czech Republic
24-27 September 2012

Abstracts

Table of Contents- Abstracts

Preliminary programme

24 Sept. 2012	
16:00	Registration
18:00	Welcome cocktail
19:00	End of registration and cocktail

25 Sept. 2012	
09:00	**Welcome address (NEA & Hosts)**
Session I: International and National programmes	
09:10	USA *P. Lyons*
09:40	NEA *Th. Dujardin*
10:00	EC *D. Haas*
10:20	France *B. Boullis and D. Warin*
10:40	*--- Coffee break ---*
11:10	Japan *TBD*
11:30	Russia *M. Kormilitsyn*
11:50	Korea *Hansoo Lee*
12:10	United Kingdom *R. Taylor*
12:30	Czech Republic *J. Uhlir*
12:50	*--- Lunch ---*
Session II: Fuel cycle strategies and transition scenarios	
14:30	Scenarios for MA transmutation *C. Coquelet (France)*
15:00	Westinghouse's Fuel Cycle Strategy *F. Franceschini (USA)*
15:30	Scenario for Closing Nuclear Power Generation *K. Nishihara (Japan)*
16:00	*--- Coffee break ---*
16:30	Systematic Comparison of Fuel Cycle Options *T. Kim (USA)*
17:00	Multinat. approach to the back-end fuel cycle *H. Noh (Korea)*
17:30	*--- Meeting adjourned ---*
19:00	*Dinner*

26 Sept. 2012	
Session II: Fuel cycle strategies and trans. scen. (cont.)	
08:30	Impact of data uncertaint. on closed fuel cycle *V. Romanello (Germany)*
09:00	The INPRO GAINS Collaborative Project *B. Dixon (USA)*
Session III: Impact of P&T on geological disposal	
09:30	Impact of MA transm. options on storage *C. Chabert (France)*
10:00	Infl. of alternat. fuel cycles on disposal uncert. *M. Nutt (USA)*
10:30	*--- Coffee break ---*
Session IV: Transm. Syst.: design, perform. and safety	
11:00	Progress of the MYRRHA project *G. van den Eynde (Belgium)*
11:30	Analysis of minor actinide incineration *B. Vezzoni (Germany)*
Session V: Pyro and aqueous separation processes	
12:00	P&T related activities in CRIEPI *M. Iizuka (Japan)*
12:30	*--- Lunch ---*
14:00	Co-processing of actinides in fuel cycles *J-P. Glatz (EC)*
14:30	The EURATOM collaborative ACSEPT project *S. Bourg (France)*
15:00	Reprocessing of spent nitride fuel *T. Satoh (Japan)*
15:30	*--- Coffee break ---*
Session VI: Transmutation fuels and targets	
16:00	MARIOS exp. on MA Bearing Blanket concept *E. D'Agata (EC)*
16:30	Qualification of MA fuels for fast reactor *N. Chauvin (France)*
Poster session (with refreshments)	
17:00	

27 Sept. 2012	
Session VI: Transmutation fuels and targets (cont.)	
08:30	Metallic fuels for actinide transmutation *R. Kennedy (USA)*
09:00	New process for dense U-Am-ox fuel fabr. *F. Lebreton (France)*
09:30	Microwave gelation *C. Cozzo (Switzerland)*
Session VII: Transm. physics, experim. and nuclear data	
10:00	Material compat. and salt chemistry control *V. Ignatiev (Russia)*
10:30	*--- Coffee break ---*
11:00	NEA group on integr. exp. for MA management *S. Okajima (Japan)*
11:30	Impact of Thorium recovery fraction *B. Petrovic (USA)*
12:00	J-Parc transmutation exp. Facility *T. Sasa (Japan)*
12:30	*--- Lunch ---*
Session VIII: Economics of P&T	
14:00	Status of the EC project ARCAS *G. van den Eynde (Belgium)*
14:30	Nuclear Energy System Cost Modeling *B. Dixon (USA)*
15:00	*--- Coffee break ---*
Summary session	
15:30	Session chair summaries
16:15	General discussion
17:00	*--- Meeting adjourned ---*

Session I -
International and National programmes

Chairs:

Status of Advanced Reprocessing and Minor Actinide Partitioning Research in the UK

Robin Taylor[1], Francis Livens[2], Andy Mount[3], Laurence Harwood[4], Chris Rhodes[5], Bruce Hanson[5]
[1]National Nuclear Laboratory, Sellafield, Seascale, CA20 1PG, U.K.
[2]School of Chemistry, University of Manchester, Manchester, M13 9PL, U.K.
[3]School of Chemistry, University of Edinburgh, West Mains Road, Edinburgh, EH9 3JJ, U.K.
[4]Department of Chemistry, University of Reading, Whiteknights, Reading, RG6 6AD, U.K.
[5]National Nuclear Laboratory, Chadwick House, Birchwood, Warrington WA3 6AE, U.K.

Abstract

The UK has a long track record in research and development (R&D) related to nuclear fuel reprocessing and in particular the PUREX process. Over the last two decades R&D has been focused towards supporting the continued operations of the two reprocessing plants at the Sellafield site, reprocessing uranium metal Magnox and uranium oxide fuels, and the development of advanced PUREX processes for future fuel cycles. Specifically, the development of single cycle reprocessing flowsheets aimed at co-processing uranium, neptunium and plutonium has been a major target. More recently, mainly through participation in European framework projects, there has been significant growth in industrial (national laboratory) and academic (university) research in minor actinide partitioning and pyrochemical processing. Some significant highlights from these European collaborations include the synthesis of a range of soft donor ligands which show extraordinary selectivity for trivalent actinides over the trivalent lanthanides and the development of a new grouped actinide extraction (GANEX) process using a combination of TODGA and DMDOHEMA ligands in the organic phase. Additional recent highlights are the UK-based MBASE and REFINE projects, focusing respectively on probing the molecular basis of aqueous separations and networking groups interested in pyrochemical actinide separations.

Progress in R&D on Pyrochemical Partitioning Technology in the Czech Republic

Jan Uhlíř
Nuclear Research Institute Řež plc,
CZ-250 68 Husinec-Řež 130, Czech Republic
E-mail: uhl@ujv.cz

Abstract

Pyrochemical partitioning represents a prominent part of the Czech R&D activity in partitioning and transmutation. The R&D activities in pyrochemistry has been focused exclusively on fluoride technologies as the original intention of the program was motivated by the Molten Salt Reactor system concept with fluoride salts based liquid fuel, the fuel cycle of which is based on pyrochemical fluoride partitioning of spent fuel. At present, the aim of the pyrochemical program is broader, in addition to MSR fuel cycle technology the utilization of the fluoride separation processes within the fuel cycles of LWR or Fast Breeder Reactors is investigated as well.

Two main fluoride partitioning technologies are under development in the Nuclear Research Institute Řež plc. The first technology devoted to the reprocessing of LWR or FBR spent fuel and to the primary processing of MSR transuranium fuel is Fluoride Volatility Method. The second technology under development is 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 Fluoride Volatility Method is focused to the development and experimental verification of a semi-pilot technology for reprocessing of advanced types of oxide, metallic and dispersed fuels from LWRs or FBRs. 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. The current research work in the area of Fluoride Volatility Method is focused to the experimental program carried out at the semi-technological line called FERDA. The experimental test program has been focused mainly to the study of flame fluorination process.

R&D on electroseparation processes from fluoride molten salt media is focused on the development of suitable electroseparation technique for partitioning of actinides from fission products in the fluoride melt media. Here the emphasis is put also on the specific aspects and requirements of on-line reprocessing technology devoted to MSR systems with thorium – uranium fuel.

The paper summarizes the results achieved in the development of pyrochemical partitioning technologies mentioned above and outlines the future activities in Czech P&T program.

Session II -
Fuel cycle strategies and transition scenarios

Chairs:

Scenarios for Minor Actinides Transmutation in the Frame of the French Act for Waste Management

C. Coquelet-Pascal, M. Meyer, M. Tiphine, R. Girieud, R. Eschbach, C. Chabert
CEA, DEN, Cadarache, F-13108 Saint-Paul-lez-Durance, France.
Tel. +33-4-42-25-77-70, Fax. +33-4-42-25-75-95, E-mail:
christine.coquelet@cea.fr

C. Garzenne, P. Barbrault
EDF R&D, France.

L. Van Den Durpel, M. Caron-Charles, B. Carlier, J-C. Lefèvre
AREVA, France.

Abstract

In the frame of the French Act for waste management, options of minor actinides (MA) transmutation are studied, based on several scenarios of Sodium Fast Reactor deployment. Basically, one of these scenarios considers the deployment of a 60 GWe SFR fleet in two steps (20 GWe from 2040 to 2050 and 40 GWe in addition from 2080 to 2100). For this scenario, the advantages and drawbacks of different transmutation options are evaluated:

- transmutation of all minor actinides or only of americium.
- transmutation in homogeneous mode (MA bearing fuel in all the core or just in the outer core) or in heterogeneous mode (MA bearing radial blankets).

Scenarios have been optimized to limit the impacts of MA transmutation on the cycle:

- reduction of the initial MA content in core in the case of transmutation in homogeneous mode to reduce the impact on reactivity coefficients.
- reduction of the number of rows of blankets and the fuel decay heat in the case of transmutation in heterogeneous mode.

The sensitivity of transmutation options to cycle parameters such as the fuel cooling time before transportation is also assessed. Thus, the transmutation of only americium in one row of radial blankets containing initially 10 wt% Am and irradiated during the same duration than the standard fuel assemblies appears to be a suitable solution to limit the transmutation impacts on fuel cycle and facilities.

A comparison of results obtained with MA transmutation in dedicated systems is also presented with a symbiotic scenario considering ADS (Accelerator Driven System) deployment to transmute MA together with a SFR fleet to produce energy. The MA inventory within the cycle is higher in case of transmutation in ADS than in case of transmutation in SFR.

Considering the industrial feasibility of MA transmutation, it appears important to study “independently” the SFR deployment and the MA transmutation. Consequently, scenarios of progressive introduction of MA options are assessed:

- beginning of MA partitioning and transmutation once the SFR fleet is totally deployed, which comes to transmuting only the MA produced by the SFR fleet at equilibrium,

- no MA transmutation at the beginning of SFR deployment, then introduction of MA transmutation in radial blankets, and finally in homogeneous mode at the SFR fleet equilibrium. A similar scenario with only americium transmutation is also considered.

Results of these progressive scenarios in terms of MA inventory in waste and in cycle are found to be intermediate between those of scenarios dedicated to each transmutation option. The MA inventory in waste is function of the date of the beginning of MA partitioning.

Westinghouse’s Fuel Cycle Strategy towards a “300-year” Nuclear Waste

Fausto Franceschini, Paolo Ferroni, Michael Wenner, Ed Lahoda, Mario Carelli
Westinghouse Electric Company, Cranberry Township, PA, USA

Abstract

Currently in the US alone, about 70,000 metric tons of Used Nuclear Fuel (UNF) are stored in fuel pools at the reactor sites or dry stored in casks, waiting for final disposal or alternative solutions. In addition to this “legacy” waste, “new” waste is continuously generated by the operating fleet. Although counting for only about 1% of the total UNF mass, the transuranic (TRU) component of UNF dominates radiotoxicity and heat load in the mid- to long-term after fuel discharge. The permanent disposal of UNF in a geological repository is perceived with scepticism by the public and part of the scientific community, primarily due to the lack of confidence in the claimed repository performance over extended periods of time. This has created a political and societal stalemate which is hampering new builds and threatens nuclear energy growth in current and potential new markets.

To resolve the stalemate and increase public confidence, Westinghouse is proposing an approach to reduce considerably the high level waste (HLW) radiotoxicity by recovering and recycling the actinides contained in the discharged fuel, while burning the TRU waste accumulated in the LWR UNF. The challenge is to develop a nuclear system that is capable of such undertaking while generating electricity in a safe and economically sound fashion. Another major technological element to obtain this goal is to develop a manufacturing and reprocessing technology for the transmutation fuel which can be scaled to an industrial level.

This paper will discuss the fuel cycle strategy and underlying technological elements which are currently under investigation at Westinghouse. The current emphasis is on a combination of LWR and Fast Reactor technology to be deployed on a properly timed schedule. LWRs are the mainstay of the commercial nuclear industry of the current century and their potential role in carrying out the transmutation mission has attractive benefits. The cost and technological risks associated to an accelerated fast reactor deployment would be avoided while allowing for the development of manufacturing and reprocessing technologies compatible with the production of transmutation fuel at an industrial scale.

In particular, Westinghouse is exploring the use of LWRs retrofitted to a reduced moderation lattice to carry out the initial phase of the transmutation. The reduced moderation lattice has harder spectrum than a regular LWR lattice thereby mitigating some of the concerns associated to multi-recycled TRU-bearing fuel. Fast reactors (FR) are also being considered and can certainly be incorporated within the fuel cycle strategy proposed. They offer the perspective of ultimate fuel cycle flexibility and fuel utilization, which together with other potential advantages would make them an unquestionable strategic asset. On the other hand FRs should be developed and introduced at the proper pace and time to allow for its technology to mature and become economically competitive.

A gradual transition to the thorium fuel cycle is also being considered as part of the Westinghouse long-term fuel cycle strategy. Thorium has some key

potential advantages, including reduced endogenous generation of higher actinides (Am, Cm atc.), better feedback coefficients, especially void reactivity coefficient, higher thermal conductivity and melting point, which can be translated into design simplification and/or improved operational and safety behaviour. However, the use of thorium presents also significant challenges and technological gaps. If these gaps can be filled, thorium could certainly play an important role in the future of the nuclear fuel cycle.

SCENARIO STUDY FOR CLOSING NUCLEAR POWER GENERATION

Kenji Nishihara, Kazufumi Tsujimoto and Hiroyuki Oigawa
Japan Atomic Energy Agency, Japan

Abstract

When we think of the end of the nuclear power generation in the future of a certain country, there will be considerable amount of nuclear material that will be no longer burnable in the commercial nuclear power reactors. Some countries may stop nuclear power generation after they operate only the light water reactor (LWR) with the use of the uranium fuel. They will have depleted uranium, plutonium (Pu) and small amount of minor actinides (MA) in the spent fuel. Others may stop after introducing a fleet of the fast breeder reactor (FBR), and they will have larger amount of Pu and MA in the FBR spent fuel than those in the LWR one. When they stop the nuclear power generation, they have to choose among directly-disposing of the spent fuels, transporting them to the other country and burning them in the dedicated transmutation systems. The first choice might be superior in economics aspect, but difficult to find and construct the repository owing to large size and possibility to recover Pu for nuclear weapon. The second one is reasonable if neighbouring countries need them, but is not available for the last country that stops the nuclear power generation. The third one is costly but effective to minimize the burden of repository.

In the transmutation strategy, Pu has to be transmuted prior to MA because of its amount and usability as the nuclear weapon. For the country without the FBR technology, Pu must be transmuted in the LWR dedicated for the transmutation in forms of the MOX fuel or the ROX fuel. The ROX fuel that stands for “rock-like oxide” fuel contains mainly Pu with less amount of 238U to reduce Pu production and to transmute about 90% of fissile Pu. The spent ROX fuel is so stable to confine radio-nuclides and it is difficult to recover Pu after disposal. Both transmutation methods in the LWR can reduce the emission of radio-nuclides to the environment and the opportunity to use Pu in the repository as the weapon. However, the potential radio-toxicity is not reduced due to remaining Pu and MA. If the transmutation systems such as fast reactor or accelerator-driven system are introduced in the nuclear fuel cycle, very small amount of Pu and MA are contained in the high-level waste, and, the potential radio-toxicity is reduced by one to two orders of magnitude.

In the present study, above mentioned transmutation methods were investigated quantitatively in terms of necessary number of transmutation systems and period after closing the nuclear power with the use of the analysis code for nuclear material flow. Moreover, the comparison of repository size and its potential radio-toxicity were made along the transmutation scenarios.

SYSTEMATIC COMPARISON OF FUEL CYCLE OPTIONS USING MASS FLOW DATA IN FUEL CYCLE DATA PACKAGES

T. K. Kim[1)], **T. A. Taiwo**[1)], **E. A. Hoffman**[1)], **R. Wigeland**[2)]
[1)] Argonne National Laboratory, U.S.A
[2)] Idaho National Laboratory, U.S.A

Abstract

The Department of Energy’s Office of Nuclear Energy (DOE-NE) has issued a Nuclear Energy Research and Development Roadmap report discussing its nuclear research, development and demonstration activities that will ensure nuclear energy’s continuing contribution as a viable energy option and its possible expansion in the United States. One of the key objectives stated in the roadmap is the development of sustainable nuclear fuel cycles that improve natural resource utilization and that provide adequate capability and capacity to manage wastes produced by the fuel cycle. To achieve this objective and enable prioritization of future activities, the DOE-NE Fuel Cycle Technologies (FCT) Program Office is conducting an evaluation and screening of nuclear fuel cycle options. As part of the evaluation and screening effort, Fuel Cycle Data Packages (FCDPs) are being developed for collecting high-quality and traceable technical data on fuel cycle options. An FCDP consists of (1) a System Datasheets file, which contains fuel cycle system information for a specific fuel cycle option, and (2) Technology Datafiles, which are wiki-style information on technologies supporting the fuel cycle option.

Currently, FCDPs for a wide-range of fuel cycle options, including once-through, limited-recycle, and continuous-recycle systems, have been partially developed. The mass flow data that are part of the system datasheets have been compared and presented in this paper, for selective fuel cycle options. Systems parameters compared in the paper include the required mass of natural uranium or thorium, utilization of natural resources, required capacity of separation facility, amount of waste, etc. A preliminary assessment of the relative performance of the fuel cycle options to a reference once-through fuel cycle option is also provided.

PROS AND CONS OF MULTINATIONAL APPROACH TO THE BACK-END FUEL CYCLE

Hyunyub Noh, Jaeyeong Park and Il Soon Hwang
Seoul National University, Republic of Korea

Abstract

In spite of Fukushima Daiichi disaster, enormous demands for nuclear energy especially in many emerging states are still existing and increasing. With steadily growing civilian nuclear power industry, needs for stable supply of nuclear fuels (front-end fuel cycle) and safe disposal of spent fuels (back-end fuel cycle) are further intensified with time. However, the related nuclear technologies such as uranium enrichment and spent fuel reprocessing have been the source of clandestine nuclear weapon materials and hence controlled duly by various national and international mechanisms in current non-proliferation regime. Multinational approaches could facilitate solutions for peaceful uses of nuclear while eliminating the risk of proliferation and terror. In this paper we have compared important proposals of multinational approaches to the nuclear fuel cycle and summarized pros and cons in implementing as a workable solution. In the case of front-end fuel cycle, bona fide efforts in implementing multinational approaches have yielded fruitful results with such as International Nuclear Fuel Bank and International Uranium Enrichment Centre. On the other hand, there is no such consensus for spent fuel recycling with continuing debates on the environment-friendliness as well as proliferation resistance. Analysis of debates on Direct Disposal or Partitioning and Transmutation (P&T) in key aspects including economy, environmental impact, and non-proliferation has led to a conclusion that advanced P&T has enough merits that warrants serious multinational approach toward demonstration and deployment. Such efforts can also provide a logical framework to maximize benefits and overcome obstacles in front. With the comparisons of outstanding options for back-end fuel cycle, practical multinational approaches have been formulated and recommended for international reviews.

IMPACT OF NUCLEAR DATA UNCERTAINTIES ON CLOSED FUEL CYCLE SCENARIOS: PRELIMINARY ASSESSEMENT

Vincenzo Romanello[1], Massimo Salvatores[2], Andrei Rineiski[1], Fabrizio Gabrielli[1]
[1]Karlsruhe Institute of Technology (KIT) – Institute for Nuclear and Energy Technologies (IKET) – Germany
[2] Commissariat à l’énergie atomique et aux énergies alternatives, France

Abstract

Nuclear energy is considered today, despite recent events, a reliable, clean and cost effective energy source. As environment protection and natural resources availability become crucial issues, nuclear research is focusing on innovative reactors and associated fuel cycles able to optimize resources exploitation and nuclear waste management. The proposed fuel cycles employ partitioning and transmutation strategies in order to minimize waste inventories. Implementation of these cycles may influence appreciably parameters such as fuel radiation activity during its fabrication, waste decay heat and radiotoxicity in a final repository. Therefore it is important to assess these parameters, which affect the technical complexity of the proposed cycle options and consequently their costs. It is also important to assess the associated uncertainties, influenced, in particular, by uncertainties in neutron cross-sections of heavy nuclides. For this purpose, different scenarios have been analyzed with the COSI code and NUCLEONICA, the latter being used for decay calculations. The first outcome of these analyses i.e. the assessment of the most important nuclei for the evaluation of doses at fuel fabrication, of decay heat at fuel discharge and in a repository, of waste radiotoxicity, etc. is presented in the paper.

Fuel Cycle Analysis Framework Base Cases for the IAEA/INPRO GAINS Collaborative Project

Brent Dixon
Idaho National Laboratory, U.S.A.
Brent.Dixon@inl.gov

Abstract

Thirteen countries participated in the Collaborative Project GAINS "Global Architecture of Innovative Nuclear Energy Systems Based on Thermal and Fast Reactors Including a Closed Fuel Cycle", which was the primary activity within the IAEA/INPRO Program Area B: "Global Vision on Sustainable Nuclear Energy" for the last three years. The overall objective of GAINS was to develop a standard framework for assessing future nuclear energy systems taking into account sustainable development, and to validate results through sample analyses. This paper details the eight scenarios that constitute the GAINS framework base cases for analysis of the transition to future innovative nuclear energy systems. The framework base cases provide a reference for users of the framework to start from in developing and assessing their own alternate systems. Each base case is described along with performance results against the GAINS sustainability evaluation metrics. The eight cases include four using a moderate growth projection and four using a high growth projection for global nuclear electricity generation through 2100. The cases are divided into two sets, addressing homogeneous and heterogeneous scenarios developed by GAINS to model global fuel cycle strategies. First, the business as usual (BAU) cases model a homogeneous world scenario with only LWRs and HWRs and no reprocessing. Next, the BAU-FR cases extend the BAU cases to include the introduction of fast reactors starting in the first half of the century and slowly replacing LWRs in the second half of the century. The rate of introduction of FRs is specified to 2050, after which they are commissioned based only on availability of plutonium for their start-up. The heterogeneous world scenario considers three separate nuclear groups based on their fuel cycle strategies, with separate non-synergistic and synergistic cases. Two of the groups, G1 and G2, are modeled to represent the existing global nuclear infrastructure, split between countries pursuing a closed fuel cycle with recycling and fast reactors (G1) and countries continuing to use a once-through fuel cycle without reprocessing (G2). The third group (G3) represents new nuclear growth and is modeled in the non-synergistic scenario as a standalone group that develops its own fuel cycle facilities. In the synergistic scenario, G3 works together with G1 and G2, obtaining fuel cycle services to support reactor deployment and operations. The framework base case analyses results show the impact of these different fuel cycle strategies while providing references for future users of the GAINS framework. A large number of scenario alterations are possible and can be used to assess different strategies, different technologies, and different assumptions about possible futures of nuclear power. Results can be compared to the framework base cases to assess where these alternate cases perform differently versus the sustainability indicators.

Session III -
Impact of P&T on geological disposal

Chairs:

Impact of Minor Actinide Transmutation Options on Interim Storage and Geological Disposal: the French Case

C. Chabert[1], D. Warin[2], JF. Milot[3], A. Saturnin[3], A. Leudet[2]
[1]Alternative Energies and Atomic Energy Commission,
CEA, DEN, Cadarache, F-13108 Saint-Paul-lez-Durance, France
Tél. +33-4-42-25-37-92, Fax. +33-4-42-25-48-49, E-mail :
christine.chabert@cea.fr
[2]CEA, DEN, Saclay, France, 3CEA, DEN, Marcoule, France

M.H. Lagrange, J.M. Hoorelbeke
ANDRA, F-92298 Châtenay-Malabry Cedex, France

Abstract

In the frame of June 28th, 2006 waste management French Act, it is requested to obtain in 2012 an assessment of industrial perspectives of partitioning and transmutation of long-lived elements. These studies must be carried out in tight connection with GENIV systems development.

The expected results must include the evaluation of technical and economic scenarios taking into account the optimization options between minor actinide (MA) transmutation processes, the interim storage and the geological disposal, based upon a systematic criteria evaluation methodology.

In this perspective, CEA asked the French waste management Agency (Andra) to assess the impact of HLW (High Level Waste) and ILW (Intermediate Level Waste) as produced by various transmutation options, on the dimensioning of the geological repository.

Three scenario studies were taken into account:

- The first scenario concerns the recycling of plutonium only in a Sodium Fast Reactor (SFR) , with the final HLW still containing the minor actinides and the fission products (PFs),
- The second scenario concerns the recycling of Pu in SFR and the MAs transmutation in MA bearing blankets; the HLW only contain FPs.
- The third scenario concerns the recycling of Pu in SFR and the transmutation of Am only in Am bearing blankets; HLW therefore includes FPs, curium and neptunium.

Andra used repository architectures similar to those employed in the Cigéo project. High level waste packages are placed in the repository cells consisting of micro-tunnels whit a diameter close to that of the waste packages. The cells are accessed via horizontal drifts located in the same plan as the cells. The cells are metal cladded in order to ensure their dimensional stability.

Results allow comparing the underground footprint and the excavated volume for each scenario; the impact of the interim storage duration (70 years or 120 years) is also assessed. An analysis of the advantages and drawbacks of transmutation options on the interim storage are performed. Solutions are proposed to optimize the footprint of the ILW and HLW repository.

Influence of Alternative Fuel Cycles on Uncertainty Associated with Geologic Disposal

Mark Nutt
Argonne National Laboratory, USA[1]

Rob P. Rechard
Sandia National Laboratories, USA

Mark Sutton, James A. Blink, H.R. Greenberg, M. Sharma
Lawrence Livermore National Laboratory, USA

Bruce A. Robinson
Los Alamos National Laboratory, USA

Abstract

Advanced nuclear fuel cycles, including those that partition and transmute key radionuclides, could potentially prove beneficial to managing high-level radioactive waste that must be stored, transported, and ultimately disposed. The potential benefits of advanced nuclear fuel cycles have been evaluated previously, focusing on potential to reduce the long-term risk or dose of geologic disposal and efficiencies that could be gained with respect to thermal management within a mined geologic repository. These efforts have sometimes claimed that the removal of key radionuclides from the waste could have an added benefit of reducing the effort required to treat uncertainty within a safety assessment. This paper explores this claim by providing a qualitative evaluation of the impact advanced nuclear fuel cycles on the treatment of uncertainty associated with geologic disposal.

The characterization of uncertainty and its treatment within a safety analysis is a fundamental aspect of evaluating the performance of geologic disposal. In this context, uncertainty includes parameter uncertainty, model form uncertainty, and the technical basis for inclusion or exclusion of specific features, events, and processes (FEPs) as part of scenario uncertainty.

A considerable amount of work has been completed worldwide regarding the treatment of uncertainty within a safety analysis for geologic disposal, but has considered only the waste streams and waste forms generated by nuclear fuel cycles currently in use. Regulators have also established requirements and guidance for treating uncertainty within the safety analysis. This evaluation leverages on this knowledge and broadens it to consider the impacts of advanced nuclear fuel cycles.

The qualitative evaluation of the impact on uncertainty from alternative fuel cycles was conducted by considering a generic list of FEPs, as derived from the NEA FEP database, applicable to the geologic disposal of high-level waste in a variety of generic geologic media (mined geologic disposal in salt, clay/shale,

---

[1] This abstract has been authored by the national laboratories operated for the US Department of Energy (DOE). Accordingly, the US Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for US Government purposes.

crystalline rock and deep borehole disposal in crystalline rock). Two aspects of impact are pertinent to the evaluation of such FEPs: (a) the effort to characterize the uncertainty, and (b) the impact on components of the safety analysis model. Both direct and indirect influences of the radionuclide inventory within the waste on each FEP were determined. An example of a direct influence would be release of key radionuclides from a degrading waste form. An example of an indirect influence would be coupled thermal-hydrologic-chemical processes where the heat generated by the waste depends on the radionuclide inventory in the waste. If no such influence is identified, it can be concluded that the uncertainty associated with that FEP is independent of the radionuclide inventory and, therefore, independent of the fuel cycle. An example of such a FEP is ground water flow far from the repository.

Session IV -
Transmutation systems: design, performance and safety

Chairs:

Progress of the MYRRHA ADS Project in Belgium.

Gert Van den Eynde, Hamid Aït Abderrahim
Belgian Nuclear Research Centre (SCK•CEN)
Boeretang 200, BE-2400 Mol – Belgium
gert.van.den.eynde@sckcen.be

Abstract

The MYRRHA project started in 1998 by SCK•CEN in collaboration with Ion Beam Applications (IBA, Louvain-la-Neuve), as an upgrade of the ADONIS project. MYRRHA is designed as a multi-purpose irradiation facility in order to support research programs on fission and fusion reactor structural materials and nuclear fuel development. Applications of these are found in Accelerator Driven Systems (ADS) and in present generation as well as in next generation critical reactors. The first objective of MYRRHA however, will be to demonstrate on one hand the ADS concept at a reasonable power level and on the other hand the technological feasibility of transmutation of Minor Actinides (MA) and Long-Lived Fission Products (LLFP) arising from the reprocessing of radioactive waste. MYRRHA will also help the development of the Pb-alloys technology needed for the LFR (Lead Fast Reactor) Gen.IV concept.

Transmutation of minor actinides can be completed in an efficient way in fast neutron spectrum facilities. Both critical reactors and sub-critical ADS are potential candidates as dedicated transmutation systems. However, critical reactors, heavily loaded with fuel containing large amounts of MA, pose reactivity control problems, hence safety problems, caused by unfavourable reactivity coefficients and small delayed neutron fraction. A sub-critical ADS operates in a flexible and safe manner even with a core loading containing a high amount of MA leading to a high transmutation rate. Thus, the sub-criticality is not a virtue but rather a necessity for an efficient and economical burning of the MA. Besides these reactor and core physics considerations, there are other parameters to consider for deciding on the most appropriate technology suitable for the large scale deployment of partitioning and transmutation (P&T) technology for the reduction of the HLW burden.

The implementation of P&T needs the demonstration of the feasibility of several installations at an “engineering” level. The respective research and development activities could be arranged in four so called “building blocks” aiming to:

A. demonstrate the capability to process a “sizable” amount of spent fuel from commercial power plants (i.e. LWR) in order to separate Pu and MA,

B. demonstrate the capability to fabricate at semi-industrial level the dedicated fuel needed to load a dedicated transmuter,

C. make available one or more dedicated transmuters,

D. provide an installation for processing of the fuel unloaded from the transmuter which can be of a different type of the one used to process initially the original spent fuel unloaded from the commercial power plants (i.e. LWR).

In this paper we will report on the progress of the MYRRHA project and its perspective of implementation in Belgium.

ANALISYS OF MINOR ACTINIDES INCINERATION ADOPTING AN INNOVATIVE FAST REACTOR CONCEPT

B.Vezzoni[1], F.Gabrielli[1], A. Rineiski[1], M. Marchetti[1], W.Maschek[1], M.Salvatores[1], A.Schwenk-Ferrero[1], V.Romanello[1], G.Forasassi[2]
[1] Karlsruhe Institute of Technology (KIT), Germany.
[2] University of Pisa (UNIPI), Italy.

Abstract

For the long term nuclear energy sustainability, the transition to a fast reactor (FRs) based fleet and the adoption of closed fuel cycles is envisaged for answering both for resources optimization and waste reduction.

The fuel cycle actually implemented is a once-through cycle based on uranium consuming reactors with thermal spectrum. In this cycle, only ~1% of the uranium extracted is used and the spent fuel is sent to disposal without reusing the fissile material (e.g. Pu239). This leads to a high demand on disposal capacities in terms of masses, radiotoxicity and heat load.

Due to specific features of fast spectrum reactors, fuels containing a fraction of Minor Actinides (MAs) can be loaded into their cores and closed (or partially closed) fuel cycles can be implemented thus providing an option for MAs transmutation. However, advantages in sustainability should not lead to a lower safety level.

Within the Collaborative Project on European Sodium Fast Reactor, CP-ESFR, reference designs for cores with oxide and carbide fuels were proposed by CEA (France). Both systems show a positive Sodium Void Reactivity Effect (SVRE) at BOL.

In the present paper, the optimization process followed for reducing the BOL SVRE is described. Only the oxide core has been considered. The main modifications are related to the axial structure by introducing a larger Na plenum (60 cm) close to the core, an absorber layer above and a lower fertile blanket. All together these attempts allow improving neutron leakages under voided conditions and therefore reducing the SVRE. In the paper, we refer to the optimized configuration as CONF2.

The low SVRE in CONF2 (about 2$ less than the reference configuration) offers an opportunity for introduction of MAs in core. Heterogeneous and homogeneous strategies have been considered within the CP-ESFR project.

In the present paper, we focus only on homogenous loadings considering the introduction of 2 and 4%wt. AmO_2 in core plus lower blanket. The results in terms of safety and fuel cycle characteristics have been compared.

The introduction of MAs implies the deterioration of safety parameters (void, Doppler, etc.) but allows the core burning its own waste.

The advantages of Am loading in terms of fuel cycle have been evaluated considering the impact on fuel cycle facilities, fuel composition evolution, radiotoxicity and heat load.

In order to assess these parameters, a simplified reference scenario representative of a country that wants to develop nuclear energy in isolation has been selected. A constant nuclear energy production in the period 2020-2200

has been defined to better underline the parameters affecting the dynamic of the transition from Light Water Reactors (LWRs) to FRs.

Only Pu and MAs actinides produced in the cycle have been considered focusing on the characteristics of the reactor in transmuting its own waste by reaching an equilibrium composition.

Neutronic analyses have been performed by means of the deterministic ERANOS code systems using JEFF3.1 data library and scenarios mentioned above have been modelled with the COSI6 code, the dynamic scenario code developed at CEA (France), widely used in Europe.

Session V -
Pyro and aqueous separation processes

Chairs:

RECENT P&T RELATED ACTIVITIES IN CRIEPI: DEVELOPMENT OF PYROCHEMICAL PROCESSING AND METALLIC FUEL TECHNOLOGIES

Masatoshi Iizuka, Takanari Ogata and Tadafumi Koyama
Central Research Institute of Electric Power Industry, Japan

Abstract

The development of innovative nuclear fuel cycle technology that has the advantages of economic and safe power generation as well as proliferation resistance is strongly expected to be an effective measure for achieving environmental sustainability and satisfying the increasing energy demand. Metallic fuel cycle technology, consisting of a metallic fuel fast reactor, pyrometallurgical reprocessing and fuel fabrication by injection casting, has been attracting increasing attention as one of the most promising nuclear fuel cycle technologies for achieving the above requirements.

From the viewpoint of partitioning and transmutation as well, the metallic fuel cycle technology has excellent advantages. The metallic fuel which has a standard composition of U-Pu-Zr incorporates very high content of the minor actinides (MA) homogeneously into itself and gives superior MA burning efficiency. In the pyrometallurgical reprocessing, no additional steps for MA recovery are needed contrary to the aqueous processes since it always accompanies the U-Pu product inevitably due to their similar thermodynamic properties.

CRIEPI started research and development on the pyrochemical processing and metallic fuel technologies at 1986 with domestic and international collaborations. Since 1994, CRIEPI and JAEA have jointly started studying the basics of actinide behaviours in molten salt and liquid metal systems, and expanded the joint study to carry out the integrated pyro-processing test and the metal fuel fabrication test for irradiation in JOYO reactor. The basic feasibility of pyrometallurgical reprocessing, such as the recovery of uranium and transuranium elements by electrorefining, has already been confirmed. Since 2009, an engineering-scale fuel cycle test as a project entrusted by Japanese government is in progress to obtain the data required for design of pyro-processing equipment for practical use. In the joint study between CRIEPI and JRC-ITU, the irradiation integrity of MA containing metal fuels up to 10 at.%BU and the recovery of MAs from both irradiated metal fuels and spent MOX fuels have been demonstrated.

Regarding the pyrochemical technology, additional advantage lies in its flexibility to accept the other type of fuels, such as oxides and nitrides. Conventional UO_2 and MOX fuels can be supplied to the pyrometallurgical treatment after reduction to metals by adoption of the electrochemical reduction technique. In the joint study with JRC-ITU, it has been demonstrated that the irradiated MOX fuels can be successfully reduced to metals by this method. After this technological achievement and the adaptability to diverse materials of various physical/chemical properties, the pyro-processing is currently under preliminary evaluation for its applicability to the treatment of the corium, mainly consisting of $(U,Zr)O_2$, formed during the accident of the Fukushima Dai-ichi nuclear power plant.

Co-processing of actinides in nuclear fuel cycles

P. Soucek, R. Malmbeck, J.-P. Glatz
European Commission, JRC, Institute for Transuranium Elements,
POBox 2340, 76125 Karlsruhe, Germany

Abstract

In view of increased discussions on the safe use of nuclear energy, also the option to reduce the burden of nuclear waste to future generations is being investigated with great interest. This includes the transmutation part where a demonstration is under development at the SCK Mol (MYRRHA project) but also the separation part developed in Europe mainly at the CEA and in the European ACSEPT program. Apart from separation of individual actinides (U, Pu, Np, Am and Cm) an alternative is the co-processing of all actinides. The latter approach represents clearly a simpler process scheme due to a reduced number of steps and a better proliferation resistance due to impure products. A major disadvantage is the need of remote handling at the re-fabrication of targets.

At ITU investigations of the co-processing options are under way both for the aqueous- and the molten salt routes. In this paper the status of both studies is being described and discussed.

ACSEPT – a successful story in actinide separation process development in EUROPE

Stephane BOURG [1]
Andreas Geist [2], **Laurent Cassayre** [3], **Chris Rhodes** [4], **Christian Ekberg** [5]
1 – CEA, Nuclear Energy Division, RadioChemistry & Processes Department,
F-30207 Bagnols sur Cèze, France
2 – KIT-INE, 3 – CNRS, 4 – NNL-UK, 5 – CHALMERS

Abstract

All along the last four years, the FP7 EURATOM Collaborative Project ACSEPT (Actinide recycling by SEParation and Transmutation) has coordinated the European Research on aqueous and pyro- actinide chemical separation processes. A clear structuration of the work and an enthusiastic collaboration between Partners has allowed significant progress in actinide separation process development.

In the field of aqueous reprocessing, four hot-test demonstrations have been carried out based on chemical systems developed in former European Projects (NEWPART, PARTNEW, EUROPART) or directly in ACSEPT. Process flowsheets are now available for the regular SANEX, the innovative SANEX, the 1 cycle SANEX and the GANEX concepts (some of them being more elaborate alternatives to reference existing processes) paving the way for further optimisation. This progress was made possible thanks to a well driven organic synthesis work. It allowed the testing of more than 150 new molecules and the selection of about 5 of them for further process development. In addition, specific issues related to advanced fuel cycles involving materials with high Pu loading or minor actinides were addressed such us MOX dissolution or co-conversion into solid forms suitable for fuel fabrication.

In the field of pyrochemical process development, two reference core of process were selected within EUROPART (the electrorefining on solid aluminium cathode in molten chloride and the liquid-liquid reductive extraction in molten fluoride/liquid aluminium). In ACSEPT, the work has been continued with a focus on key process steps such as the exhaustive electrolysis allowing a quantitative recovery of the transuranics after the electrorefining or the actinide back extraction from an aluminium alloy, crucial step for the two reference processes. In addition, a specific effort has been allocated to the head-end step (oxide pre-reduction, thermal treatment), the salt recycling (chloride salt purification by precipitation, fluoride salt purification by distillation) and the specific salt waste conditioning.

This experimental work has been completed by cross cutting studies, such as system studies, scale up studies, corrosion studies and online process monitoring developments for molten salts. The MARIOS irradiation performed in FAIRFUELS was also designed in ACSEPT to allow a better schedule of this long run experiment.

In parallel, one of the major successes of ACSEPT has also been its investment in people with the funding of two post-doctoral positions and of a dozen of student exchanges between partners allowing these students to broaden their field of competences and to increase collaboration between partners. Specific workshops and seminars were also organised to allow a better exchange between partners.

At one week of the end of the project, this paper will highlight the main achievements of ACSEPT and will identify and propose tracks for future developments meeting the requirements of EURATOM work-program. Based on the successful work performed in ACSEPT, a new project proposal has been submitted to EC that would allow the community to optimise the selected separation processes and demonstrate and increase their safety on sound scientific bases.

REPROCESSING OF SPENT NITRIDE FUEL BY CHEMICAL DISSOLUTION IN MOLTEN SALT – RESULTS ON PLUTONIUM NITRIDE CONTAINING INERT MATRIX MATERIALS–

Takumi Satoh, Kenji Nishihara, and Masahide Takano
Japan Atomic Energy Agency, Japan

Abstract

Nitride fuel cycle for the transmutation of long-lived minor actinides (MA) has been developed in Japan Atomic Energy Agency (JAEA) under the double-strata fuel cycle concept. The transmutation system proposed by JAEA is a Pb–Bi cooled sub-critical accelerator driven system (ADS) with ^{15}N-enriched Pu-MA mixed nitride fuels containing about 70 mol% ZrN as inert matrix material. In the currently-proposed pyrochemical process of the spent nitride fuel, actinides are dissolved in LiCl-KCl eutectic melts and selectively recovered into liquid Cd cathode by molten salt electrorefining. The recovered actinides are converted to nitrides by the nitridation-distillation combined method, in which the Cd alloys containing actinides are heated in nitrogen gas stream. However, this process has some disadvantages such as (1) the decrease in the anodic dissolution rate of actinide nitride by addition of the inert matrix materials, (2) the generation of the nitride products with different composition: the product from electrorefining process and that from the processing of the anode residues, and (3) treating large amount of Pu and MA including the inventory of them in the electrorefiner.

In the present study, the innovative process combining the chemical dissolution of spent nitride fuel into molten chloride and the multi-stage counter current extraction of actinide elements from the molten chloride media with liquid Li-Cd alloy is newly considered. In the chemical dissolution of the spent nitride fuel, the actinide nitrides are dissolved into molten LiCl-KCl eutectic salt as chlorides by chemical reaction with the oxidizing agents such as $CdCl_2$. The residue materials are processed into a metal waste form in the flow diagram. The molten salt containing actinide chlorides is transferred to the multi-stage counter current extraction of actinide elements from the molten chloride to liquid Cd using Li as a reductant. The actinides recovered in Cd are converted to nitrides by the nitridation-distillation combined method. The major advantages of this process are (1) the improvement in processing rate, (2) the homogenization of the composition of the nitride products, and (3) the reduction of the amount of actinide elements treated in the reprocessing process. In the presentation, the experimental results of the chemical dissolution of plutonium mononitride containing the inert matrix materials (ZrN and TiN) are to be introduce.

This study is the result of “Development of the reprocessing of spent nitride fuel by chemical dissolution in molten salt” carried out under the Strategic Promotion Program for Basic Nuclear Research by the Ministry of Education, Culture, Sports, Science and Technology of Japan.

Session VI -
Transmutation fuels and targets

Chairs:

Preliminary results of the MARIOS experiment on Minor Actinide Bearing Blanket concept.

E. D'Agata[1], P.R. Hania[2], S. Bejaoui[3], C. Sciolla[2], T. Wyatt[2], M.H.C. Hannink[2], N. Herlet[4], A. Jankowiak[5], F.C. Klaassen[2], J-M. Bonnerot[3]
1) European Commission, Joint Research Centre, Institute for Energy – P.O. Box 2, 1755 ZG Petten, The Netherlands. Tel.: 00 31 224 56 5117; Fax.: 00 31 224 5627; e-mail: elio.dagata@ec.europa.eu
2) Nuclear Research and Consultancy Group, P.O. Box 25 1755 ZG Petten, The Netherlands
3) Commissariat à l'Energie Atomique, DEC CEA-Cadarache, 13108 St. Paul lez Durance Cedex, France
4) Commissariat à l'Energie Atomique DTEC CEA Marcoule, 30207 Bagnols sur Cèze cedex, France
5) Commissariat à l'Energie Atomique DEN DANS DMN SRMA LC2M, 91190 Gif-sur-Yvette, France

Abstract:

Americium is a strong contributor to the long term radiotoxicity of high activity nuclear waste. Transmutation by irradiation in nuclear reactors of long-lived nuclides like 241Am is, therefore, an option for the reduction of radiotoxicity, residual power packages as well as the repository area. One of the most attractive possibilities to burn MA, is represented by the Minor Actinides Bearing Blanket (MABB) concept. In this option, MA are diluted in a UO2 matrix and irradiated for a long time (4,100 EFPD) into radial blankets at the periphery of the outer core of a sodium fast reactor. The MARIOS irradiation experiment is the latest of a series of experiments on americium transmutation (e.g. EFTTRA-T4, EFTTRA-T4bis, HELIOS). Moreover, the MARIOS experiment, together with the DIAMINO experiment, which is under preparation in the OSIRIS reactor in France, is dealing with the study of the in-pile behaviour of UO2 containing Minor Actinides (MAs) in order to gain knowledge on the role of the microstructure and of the temperature on the gas release and on fuel swelling for the MABB concept. The MARIOS experiment is carried out in the framework of the 4-year project FAIRFUELS of the EURATOM 7th Framework Programme (FP7).

The MARIOS experiment is conducted in the HFR (High Flux Reactor) in Petten (The Netherlands) and started in March 2011. It has been planned that the experiment will last 11 cycles, corresponding approximately to 11 months.

This paper covers the description and the objectives of the experiment, as well as the preliminary result in term of first assessment and temperature recording.

Qualification of Minor Actinide fuels for fast reactor: fuel design and feedback of irradiation results.

N. Chauvin, M. Phelip, JM Bonnerot, S. Bejaoui.
Centre de Cadarache, DEC/SESC bat 151,
13108 SAINT PAUL LEZ DURANCE, FRANCE

Abstract

Two different modes exist for minor actinide transmutation: homogeneous route and heterogeneous route (UO2 or inert matrix support).

After twenty years of R&D focused on technical feasibility, we can make an analysis based on the main outcomes obtained for transmutation fuel to analyze where we are in the fuel qualification process.

Maturity of each transmutation fuel may be evaluated with a qualification scale. This scale is used for several types of fuel up to industrialization stage.

We have attempted to apply this method to our fuels.

The result is an evaluation of the work to be done in the coming years.

This will be followed by an adjustment of R&D programme until final qualification step with different items: fabrication process, fuel element and sub-assembly design, properties measurements, behaviour modelling and irradiation programme. The irradiation programme shall include analytical, semi-integral and integral irradiations, in MTR or in SFR prototypes in normal conditions as well as for transients and accidental conditions.

It’s also necessary to investigate all constraints coming from others analysis, mainly: scenario results, fuel cycle capabilities, reactor limits and others impacts (transport, repository, …) in order to improve the transmutation fuel design.

THE DEVELOPMENT OF METALLIC FUELS FOR ACTINIDE TRANSMUTATION

J. Rory Kennedy
Idaho National Laboratory, USA

Abstract

Idaho National Laboratory (and its predecessor Argonne National Laboratory-West) has a long tradition in studying the properties and behavior of metallic fuels for performance in fast reactor applications. Over the past 10+ years, efforts have been specifically directed towards closing the fuel cycle and studying the fabrication, properties, and reactor performance of (U,Pu,Np,Am)Zr alloy systems. Uranium free fuel compositions intended for accelerator based transmutation technologies, uranium bearing fuel compositions intended for fast reactor based transmutation technologies, and fuel compositions including fission product lanthanide element carryover from the molten salt electrochemical separations process ("pyro-processing") have been studied to date. Fabrication technologies have been developed and issues such as americium volatility addressed to success. The pre-irradiation characterization of the alloys including phase diagram and thermophysical property measurements, microstructure, room temperature as-cast phase and thermal cycled phase formation, variable high temperature XRD phase identification, transition temperatures including melting points, enthalpies of transition and heat capacity by DTA and DSC, thermal expansion by dilatometry or TMA, thermal diffusivity, and thermal conductivity. Post-irradiation examination of low and high burnup fuel samples is proceeding with results limited to date to non-destructive examination (NDE) and preliminary optical microscopy (metallography). Of particular interest is the recent application of advanced measurement techniques to the fuels, in both pre- and post-irradiation studies, and the use of these results in conjunction with fuel modeling and simulation efforts. Studies to date include employment of a focused ion beam (FIB) on irradiated fuels to create lamella for EBSD analysis to experimentally reveal grain orientation in the irradiated fuel, which can then be input to the computational effort to investigate the effect on fission gas migration. These type studies will be overviewed as well as possibly other advanced instrumentation studies on transmutation fuels from atom probe tomography, 50 μm spatial resolution thermal diffusivity, micro-focus X-ray diffraction, and electron probe microanalysis.

A NEW PROCESS FOR DENSE U1-xAmxO2±δ FUEL FABRICATION BY CONVENTIONAL SINTERING

Florent Lebreton, Denis Horlait, Thibaud Delahaye
CEA Maroule, DEN/DTEC/SDTC/LEMA, Bagnols-sur-Cèze, France

Abstract

In the frame of minor actinides, and more specifically americium, recycling via heterogeneous transmutation in FNR, MABB (Minor Actinide Bearing Blankets) fuel fabrication processes are currently being developed and optimised. Until now, these processes are based on those commonly used to prepare mixed-oxide fuels (such as U1 xPuxO2). They are thus based on a high temperature reactive sintering between two (at least) single-oxide (such as UO2+δ, AmO2-δ) precursors which can lead to defects (such as porosity or chemical/structural inhomogeneity) discrepant with requirements for such fuels. These defects are due to the competition between the two main phenomena constituting a reactive sintering, namely the solid solution formation, and the densification; the latter being inhibited as long as the former is not complete. In the case of U1 xAmxO2±δ MABB fuels, the large difference in diffusion coefficient and oxygen potential between uranium and americium oxides renders the use of reactive sintering quite challenging for the fabrication of homogeneous and dense (d > 94% TD) U1-xAmxO2±δ MABB fuels.

To overcome these limitations a new process, called UMACS (Uranium / Minor Actinide Conventional Sintering), has recently been developed at the LEMA (CEA Marcoule). This process is based on the separation of the two above-mentioned phenomena. Firstly, a solid state route is employed to synthetize a solid solution: a homogeneous powder mix of single dioxide is prepared, pelletized and heat treated. The thermal cycle and the atmosphere are chosen to favour the formation of the intended U1-xAmxO2±δ oxide solid solution. A recent in-situ study on UO2+δ/AmO2-δ interaction allows specifying the required thermal cycle. The atmosphere is composed of an Ar/H2/H2O mixture to avoid any americium sublimation while maintaining uranium oxide in the fluorite-type-structure dioxide form. Secondly, a U1-xAmxO2±δ powder is produced from those heat-treated pellets by the use of a grinding step. As-obtained monophasic powder is thus suitable for a conventional sintering. The solid solution formation/densification competition being avoided, the sintering is not impeded and highly dense samples can finally be achieved, without a significant amount of open porosity. As all the americium is already integrated in the solid solution, its sublimation is prevented, which allows varying the atmosphere in order to adjust the O/M ratio of the fuel.

The feasibility and reproducibility of this process will be exemplified by the recent fabrication and characterisation of highly dense U1 xAmxO2±δ (x = 0.075; 0.15; 0.2) MABB fuels using the UMACS process.

METHODS OF ADVANCED WASTE CONDITIONING BY MICROWAVE INTERNAL GELATION: SET UP DEVELOPMENT AND MODELING

C. Cozzo, M. Cabanes-Sempere, M. A. Pouchon
NES, Paul Scherrer Institut, 5232 Villigen, Switzerland

Abstract

Based on closed packed microspheres produced by gelation, the Sphere-pac is a promising concept for transmutation of minor actinides in fast reactors [1]. In the case of internal gelation, the chemical reaction is triggered by a temperature increase within aqueous droplets. Since microwaves provide a fast and volumetric heating, a set up is developed in PSI where the microspheres undergo gelation as they cross the electromagnetic field generated inside a cavity [2].

In this work the current set up of the particle production unit is presented. The components were selected and mounted in such a way that the production can be remotely operated. An automation program is being developed in order to optimize the critical parameters during operation. This will allow a safe and easy control of the equipment when the cavity is placed in a glovebox with radioactive materials.

The described unit has been used to heat water and cerium solution droplets, the latter element being considered as a surrogate for active materials. The obtained results are reported and discussed here. To support the experimental data, a modelling of the electromagnetic field generated in the cavity has been developed. The perturbation of the field caused by the presence of a droplet and the amount of absorbed energy can be simulated. These experimental and theoretical studies enable the optimization of the microwave cavity design and the determination of the needed power for the production of fuel microspheres.

[1] M. A. Pouchon, G. Ledergerber, F. Ingold, and K. Bakker. Sphere-Pac and VIPAC Fuel, volume 3. Comprehensive Nuclear Materials, Elsevier, Amsterdam, 2012.

[2] Maria Cabanes-Sempere, Cedric Cozzo, Sebastien Vaucher, Jose M. Catala-Civera, and Manuel A. Pouchon. Innovative production of nuclear fuel by microwave internal gelation - heat transfer model of falling droplets. Prog. Nucl. Energ., 57:111–116, 2012.

Session VII -
Transmutation physics, experiments and nuclear data

Chairs:

MATERIALS COMPATIBILITY AND SALT CHEMISTRY CONTROL: CASE FOR MOLTEN SALT ACTINIDE RECYCLER & TRANSMUTER

Victor Ignatiev, Valery Afonichkin, Olga Feynberg and Aleksandr Surenkov
NRC-Kurchatov Institute, Russia

Abstract

Last decade R&D on Molten Salt Reactor (MSR) is focused of fast spectrum concepts which have been recognized as a long term alternatives to solid fuelled fast neutron reactors with attractive features (very negative feedback coefficients, smaller fissile inventory, easy in-service inspection, simplified fuel cycle). Single and two fluid Molten Salt Actinide Recycler & Transmuter (MOSART) designs without and with fertile materials support are now available for for long lived actinides burning and new fuel breeding. It was demonstrated that the MOSART system without graphite moderator can operate with different loadings and make up based on transuranium elements from used LWR fuel without Th support as special Li,Be/F actinide burner (for spent LWR fuel with Pu/(Np+MA) ratio down to 1.5 and with Th support as self-sustainable system (CR=1) or even as a breeder (CR>1). It seems that for transition to more poor fuel feedings on the base of MA only, molten fluoride solvents with higher solubility of MAF_3 are necessary (e.g. LiF-NaF-KF eutectic fuelled by 7.6 mole % of MAF_3). But this will need very high (much more than 30 t of MA) loading to get criticality.

Selection of the salt composition strongly depends on the specific design application. For MOSART design, materials selection in primary circuit is a very important issue. This paper summarizes results of an experimental investigation conducted recently to understand the mechanism and to develop a means of controlling uniform corrosion and tellurium embrittlement in the Ni – base alloys.

The addition of a chromium telluride to salt can be used to provide small partial pressures of tellurium simulating a reactor environment where tellurium appears as a fission product. Tellurium corrosion of Ni-based alloys in stressed and unloaded conditions studies was tested at temperatures up to 750°C with measurement of the salt redox potential in LiF-(NaF)-BeF_2 and LiF-(BeF_2)-ThF_4-UF_4 fuel salts in the range of U4+/U3+ ratios from 1 till to 500. Following Hastelloy N-type modified alloys: HN80M-VI with 1.5% Nb, HN80MTY with 1% Al and MONICR with about 2% Fe were used for the study in the 15LiF-58NaF-27BeF_2 corrosion facilities. Materials investigated in molten 75LiF-5BeF_2-20ThF_4-UF_4 mixture fuelled by 2 mole% UF_4 included, in addition to mentioned above, high temperature 77Ni-7Cr-10Mo-6W and 65Ni-28W-7Cr alloys.

The intergranular cracking (IGC) produced in Hastelloy N (or HN80M) -type alloys when exposed to this chromium telluride-salt mixture can be reduced by adding niobium or aluminum to the Ni base alloy or by controlling the oxidation potential of the salt in the reducing range. It was shown that both Re and Y additions only slightly increase the alloy’s resistance to tellurium cracking. The alloy doped with Nb alone significantly increases IGC resistance. The alloy containing both Ti and Nb did not provided required resistance to tellurium corrosion. Addition of Mn gives a significant increase in alloy resistance to tellurium IGC. HN80MTY alloy is the most resistant to tellurium IGC of Ni-base alloys under study.

After materials exposure in the fuel salt with the [U(IV)]/[U(III)] ratio from 1 to 100 there was revealed no traces of tellurium intergranular cracking on specimens surface. Te IGC was found on tested alloys only after exposure in fuel salt with [U(IV)]/[U(III)] = 500. For each of the tested alloys the intensity of IGC was essentially lower in unstressed state than in stress condition.

Study on deuterium permeation through nickel-based HN80MTY and EM721 alloys is also carried out. Temperature dependences of deuterium solubility, coefficients of permeability and diffusion in alloys were built.

The practical realization of the system under consideration highly depends on the further experimental investigations of the chemical and physical properties for the chosen fuel / blanket salts and metallic materials. The status of technology and planned development activity is discussed in details.

SUMMARY OF OECD/NEA/NSC EXPERT GROUP ON INTEGRAL EXPERIMENTS FOR MINOR ACTINIDE MANAGERMENT

Shigeaki Okajima 1), Philippe Fougeras 2), Choong-Sup Gil 3), Georgios Glinatsis 4),
Jim Gulliford 5), Osamu Iwamoto 1) , Robert Jacqmin 2), Yury Khomyakov 6),
Anatoly Kochetkov 7), Mikhail V. Kormilitsyn 8), Richard Mcknight 9), Yoshiyuki Nemoto 5),
Giuseppe Palmiotti 10), Gregory Perret 11), Vincenzo Romanello 12), Kazufumi Tsujimoto 1),
Guido Vittiglio 8), Akifumi Yamaji 5)
1) JAEA, Japan, 2) CEA, France, 3) KAERI, Korea, 4) ENEA, Italy, 5) NEA, OECD,
6) IPPE, Russia, 7) SCK-CEN, Belgium, 8) RIAR, Russia, 9) ANL, USA, 10) INL, USA,
11) PSI, Switzerland, 12) KIT, Germany

Abstract

To establish a reliable fuel cycle together with managing safely the radioactive wastes is inevitable in pursuing a sustainable utilisation of nuclear fission energy. One of the key subjects to realize it is the appropriate management of minor actinides (MA), such as neptunium, americium and curium in the spent fuel. In particular, the necessity to manage MA becomes more obvious when plutonium is used as MOX fuel on a large scale in power reactors, both light-water reactors and fast reactors, since more accumulation of americium and curium will be predicted in the spent fuel. One effective way to manage MA is to transmute them in nuclear systems; fast reactors and accelerator-driven subcritical systems. The transmutation of MA, however, is not easy. From a viewpoint of the reactor neutronics, the loading of MA generally affects the reactor characteristics, such as the coolant void reactivity, the Doppler coefficient, and the burn-up effect.

The detailed design of transmutation systems with reliable accuracy and the precise prediction of the composition of the spent fuel are difficult since the accuracy of MA nuclear data is not sufficient yet. This is because there is a lack of experimental data for MA. It should be remembered that nuclear data of the major actinides, such as U-235, U-238 and Pu-239, have been improved for more than 50 years, based on a huge number of differential and integral experiments, using accelerators, critical facilities and experimental reactors.

The results of the integral experiments have been used to validate the nuclear data. Recently the covariance data in the nuclear data libraries have been introduced to estimate the uncertainty in design study of the nuclear system. The integral experimental data together with the sensitivity analysis have become more effective to improve the design accuracy. The integral experiments on MA, however, are much more difficult than those on the major actinides because of restrictions at the facilities, the difficulty of sample preparation, the necessity to improve measurement techniques to reduce the influence of background radiation, and so on.

Considering these situations, it is requested that a critical review of integral experiments for validating MA nuclear data should be undertaken with appropriate quality assurance for MA transmutation in the nuclear systems. The

Nuclear Science Committee in OECD/NEA accepted the establishment of the Expert Group (EG) on Integral Experiments for Minor Actinide Management in 2008.

The EG consisted of 19 members from 10 countries and OECD. The meetings were held 7 times between Sept. 2009 and Sept. 2012. The members discussed about the following subjects; (1) Requirement of nuclear data for MA management (including evaluation of target accuracy, comparison of uncertainty analysis results among nuclear data libraries and identification of important nuclear data), (2) Reviewing existing integral data (including review of MA sample amount used/required in experiment), (3) Specification of missing experimental work to be required, (4) Identifying and considering possible solutions to the bottlenecks, such as availability of MA sample and experimental facilities, and (5) Proposal of action program for international cooperation.

The summary of these activities and the results will be shown.

IMPACT OF THORIUM RECOVERY FRACTION ON LONG TERM RADIO-TOXICITY OF A THERMAL SPECTRUM SYSTEM HIGH LEVEL WASTE

L. Michael Huang, Bojan Petrovic
Georgia Institute of Technology, Atlanta, GA, USA
Fausto Franceschini
Westinghouse Electric Company, Cranberry Township, PA, USA

Abstract

The paper examines the use of thorium-bearing fuel in a thermal spectrum system with reprocessing. Due to inherently reduced generation of higher actinides, thorium has well-known potential to reduce the long-term radiotoxicity of nuclear waste. Most of such analyses assume a relatively high recovery fraction (e.g., 99% or 99.9%), similar to that expected in UREX-type processes. However, Thorex or other separation processes considered for thorium-bearing fuel may in fact have a significantly lower recovery fraction. This paper examines the impact of the reduced recovery and different partitioning schemes (with respect to Np and Pu) on the radio-toxicity of actinide inventory in high level waste generated by thorium-bearing fuel.

DESIGN OF J-PARC TRANSMUTATION EXPERIMENTAL FACILITY

Toshinobu Sasa, Kazufumi Tsujimoto, Hayanori Takei, Shigeru Saito, Hironari Obayashi, Kenji Nishihara, Takanori Sugawara, Hiroki Iwamoto, and Hiroyuki Oigawa
Japan Atomic Energy Agency, Japan

Abstract

After the Fukushima accident, management of spent fuel got many attentions and then, reduction of the radioactive wastes becomes increasingly important. Partitioning-Transmutation technology using fast reactors or accelerator-driven systems (ADS) is noted at re-evaluation of the national fuel cycle policy discussed by Atomic Energy Commission of Japan. To realize the transmutation of minor actinides and long-lived fission products, basic studies for various research fields are needed and therefore, equipments to obtain the experimental data using spallation target and minor actinides are required. In the framework of the J-PARC project, Japan Atomic Energy Agency (JAEA) has been promoted to construct the experimental facility for transmutation systems and performed design of Transmutation Experimental Facility (TEF). The facility consists of two buildings, Transmutation Physics Experimental Facility (TEF-P) to perform critical experiments using minor actinide bearing fuels and ADS Target Test Facility (TEF-T) for irradiation of various structural material candidates in flowing lead-bismuth environment. According to the latest time schedule, TEF-T will be constructed rather faster than licensing of TEF-P to facilitate the construction. TEF-T will be operated within 5 years and TEF-P needs another 5 years to finish construction, after the budget approbation by government. Based on this time schedule and facility layout, concepts of the facility, namely the TEF-T should be revised to accept proton beam up to 400MeV-133kW. Sealed-annular tube type spallation target filled by lead-bismuth eutectic alloy is considered for both low-power proton irradiation target and full stop length target for proton/neutron simultaneous irradiation. Both targets were designed to simulate the operating condition of actual lead-bismuth cooled ADS transmutor (800MWt). The temperature range of the lead-bismuth can be set around 300 – 500 degree centigrade and systematic data accumulation is planned for various structural materials and irradiation damage rate indicated by DPA (displacement per atom) at various operation temperatures under the structural stressed environment. Even the sample is limited in very small size, the annual irradiation rates can be set to several tens DPA. TEF-P is designed reflecting the structure of existing critical assembly FCA located in JAEA/Tokai site to take over many experimental data and experiences at FCA. Required amount of minor actinides to perform the significant critical/subcritical experiments are specified and is reflected to the design of critical assembly itself. Remote devices and related facility layout are also designed to handle the minor actinide bearing fuels safely. In the presentation, latest status for TEF construction and experimental needs discussed by atomic energy society of Japan will be introduced.

Session VII -
Economics of P&T

Chairs:

Status of the EC-FP7 project ARCAS: Comparing the economics of Accelerator Driven Systems and Fast Reactors as Minor Actinide burners

Gert Van den Eynde[1], Vincenzo Romanello[2], Tim Abram[3], Colin Zimmerman[4], Aliki van Heek[5]
[1]SCK•CEN, Belgium
[2]KIT, Germany
[3]University of Manchester, United Kingdom
[4]NNL, United Kingdom
[5]NRG, The Netherlands

Abstract

The ARCAS project aims to compare, on a technological and economical basis, Accelerator Driven Systems and Fast Reactors as Minor Actinide burners. In order to be able to perform this comparison, one needs to have a sufficient idea on the expected minor actinide stream for a reasonably large nuclear park. The assessment of this stream was the subject of the first work package. A literature study of several previous projects focusing on fuel cycle scenario analyses was performed together with an analysis of the current legal framework on spent fuel management and a review of current fuel reprocessing and fabrication techniques; some relevant data were obtained by some further elaboration of available simulations. This resulted in a reference minor actinide mass flow (a lower and a higher boundaries were estimated) and isotopic vector that can be expect based on the first PATEROS (EC-FP6) reference scenario.

In parallel, work packages 2 and 3 are establishing the reference Fast Reactor and Accelerator Driven System to be used in the ARCAS project. The main concern here is the maximal loading (from the point of view of safety and operations) of minor actinides in the core of either system. A brief overview of open studies has been done with a special emphasis on the EU project CP-ESFR (3600 MWth SFR concept), for which a set of activities are being directed towards MA transmutation. For the ADS, the EFIT design (EC-FP6 IP-EUROTRANS) has been selected by work package 3 as the reference case. A parametric study on relevant safety parameters (delayed neutron fraction, void effect) has been performed as a function of the minor actinide loading using the reference minor actinide vector from work package 1.

The fourth work package is responsible for a schematic design of the fuel reprocessing facilities and fuel fabrication facilities in support of the fuel cycle involving the fast reactors and accelerator driven systems. One critical component, and often overlooked in other studies, is the transportation issue: the cost and safety of the transportation of fuel with (high) minor actinide content.

Finally, the last work package has to gather all information from the other work packages in order to able to present a comparison between the two options of fast reactors or accelerator driven systems. With the number of units needed per GWe of LWR installed and the investment cost of a transmutation unit, the investment cost per GWe is determined. For selected nuclear evolution scenarios, the total investment cost needed for transmutation can be determined. Also, the total generating costs are compared, giving an answer to

the question on how much the MA transmutation would add to the cost of kWh. These costs would include both the investment, operational and fuel cycle costs. The fuel cycle costs consist of all the parts of the closed cycle, including reprocessing and fuel/target fabrication.

Nuclear Energy System Cost Modeling

Francesco Ganda – Idaho National Laboratory
Brent Dixon – Idaho National Laboratory

Abstract

The U.S. Department of Energy’s Fuel Cycle Technologies (FCT) Program is preparing to perform an evaluation of the full range of possible Nuclear Energy Systems (NES) in 2013. These include all practical combinations of fuels and transmuters (reactors and sub-critical systems) in single and multi-tier combinations of burners and breeders with no, partial, and full recycle. As part of this evaluation, Levelized Electricity Unit Cost (LEUC) ranges for each representative system will be calculated. To facilitate the cost analyses, the 2009 Advanced Fuel Cycle Cost Basis Report is being amended to provide up-to-date cost data for each step in the fuel cycle, and a new analysis tool, NE-COST, has been developed. This paper explains the innovative “Island” approach used by NE-COST to streamline and simplify the economic analysis effort and provides examples of LEUC costs generated.

The Island approach treats each transmuter (or target burner) and the associated fuel cycle facilities as a separate analysis module, allowing reuse of modules that appear frequently in the NES options list. For example, a number of options to be screened will include a once-through uranium oxide (UOX) fueled light water reactor (LWR). The UOX LWR may be standalone, or may be the first stage in a multi-stage system. Using the Island approach, the UOX LWR only needs to be modeled once and the module can then be reused on subsequent fuel cycles.

NE-COST models the unit operations and life cycle costs associated with each step of the fuel cycle on each island. This includes three front-end options for supplying feedstock to fuel fabrication (mining/enrichment, reprocessing of used fuel from another island, and/or reprocessing of this island’s used fuel), along with the transmuter and back-end storage/disposal. Results of each island are combined based on the fractional energy generated by each islands in an equilibrium system.

The cost analyses use the probability distributions of key parameters and employs Monte Carlo sampling to arrive at an island’s cost probability density function (PDF). When comparing two NES to determine delta cost, strongly correlated parameters can be cancelled out so that only the differences in the systems contribute to the relative cost PDFs. For example, one comparative analysis presented in the paper is a single stage LWR-UOX system versus a two-stage LWR-UOX to LWR-MOX system. In this case, the first stage of both systems is the same (but with different fractional energy generation), while the second stage of the UOX to MOX system uses the same type transmuter but the fuel type and feedstock sources are different. In this case, the cost difference between systems is driven by only the fuel cycle differences of the MOX stage.

Posters -
Session II

SCENARIOS FOR FAST REACTOR DEPLOYMENT WITH PLUTONIUM RECYCLING

C. Coquelet-Pascal, M. Meyer, R. Girieud, M. Tiphine, R. Eschbach, C. Chabert
CEA, DEN, Cadarache, F-13108 Saint-Paul-lez-Durance, France.
Tel. +33-4-42-25-77-70, Fax. +33-4-42-25-75-95, E-mail:
christine.coquelet@cea.fr
C. Garzenne, P. Barbrault
EDF/R&D, France.
L. Van Den Durpel, M. Caron-Charles, B. Carlier, J-C. Lefèvre
AREVA, France.

Abstract

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. These reactors will be initially fuelled with plutonium coming from spent MOX fuels, the breeding being adjusted according to energy needs. In this context, scenarios of Sodium cooled Fast Reactor deployment in progressive replacement of the French PWR fleet are assessed. Different chronologies are considered to evaluate the capability of SFR to renew a 60 GWe PWR fleet and the sustainability of this deployment in function of plutonium availability. The first scenario considers the deployment of SFR in two steps with 20 GWe from 2040 to 2050 and 40 GWe in addition from 2080 to 2100. The impact of a SFR deployment delayed in 2080 with 60 GWe deployed from 2080 to 2110 is assessed in a second scenario. In both cases, it is possible to deploy a 60 GWe SFR fleet at the end of the century, either by reducing the minimum cooling time before reprocessing of SFR fuel from 2080, or by adding fertile blankets to SFR.

The sensibility of scenarios to the SFR core design has been evaluated by considering different SFR types:

- the SFR V2B with a homogeneous core design;
- the CFV with a new heterogeneous core design with a significant gain on the sodium void effect.

The feasibility of SFR deployment is not affected by the change of core design and impacts on facilities are limited.

In addition to these scenarios with SFR deployed to replace the PWR fleet, scenarios of symbiotic fleet, in which a 60 GWe PWR fleet is maintained while SFR are progressively introduced from 2040 depending on the plutonium produced by the PWR fleet, can be envisaged.

The first evaluation focuses on the maximal achievable installed SFR power using plutonium in PWR spent fuel. Both cases of PWR fleet with and without MOX fuel are evaluated. The halt in 2040 of plutonium recycling in MOX PWR fuel allows indeed to deploy an additional 50 GWe of SFR in 2150.

The second part presents different key drivers to modulate the SFR deployment:

- increasing the PWR spent fuel cooling time to slow down the introduction of first SFR;

- increasing the SFR spent fuel cooling time to reduce the rhythm of SFR deployment;
- sizing spent fuel reprocessing capacities to realistic values.

These studies allow to identify configurations in which spent fuel storage are stabilized to values consistent with current values, in particular in the case of a PWR fuel cooling time fixed to 20 years or in the case of limited reprocessing capacities.

Advanced Fuel Cycle Simulator Description

Brent Dixon – Idaho National Laboratory
Katy Huff – University of Wisconsin

Abstract

This paper describes the major functions and use cases of the Advanced Fuel Cycle Simulator (FCS). The FCS is the next generation fuel cycle analysis tool that is in initial stages of development as the Grand Challenge for the Systems Analysis Campaign of the United States Department of Energy's (DOE) nuclear fuel cycle technologies program (FCT). The FCS will dynamically simulate the commercial nuclear fuel cycle at the discrete facility level with isotopic resolution. It seeks to be a versatile, open-platform effort, supporting users and developers at levels of detail appropriate for a broad and varied stakeholder base interested in a range of analysis types. Intended users include government laboratories, universities, and industry groups. The paper identifies analysis missions and summarizes the functional requirements of a comprehensive, top-level fuel cycle simulation code more fully described in the FCT report "Draft Next Generation Fuel Cycle Simulator Functions and Requirements Document". The report also proposes a design approach to complement currently available fuel cycle simulation capabilities. The FCS is intended to provide a supportive framework for research across national, institutional, and disciplinary boundaries. One of the primary missions of the FCS will be to improve understanding of advanced fuel cycle performance and the interactions of fuel cycle technologies existing and under development. This includes modeling of the impacts of advanced reactors, separations and recycling options on resource sustainability, economics, safety, security, proliferation risk, waste streams, waste form options and disposal system performance. As of the writing of this abstract, DOE has initiated a three-year effort to develop the FCS via a Nuclear Energy University Program (NEUP) grant to a consortium of four universities, led by the University of Wisconsin. The grant includes development of the core FCS functions by modifying and maturing the object-oriented CYCLUS code in an open source environment, reconfiguring the code for cloud computing so that it can be delivered on anything from an engineering workstation to a tablet computer, developing interface capabilities for different levels of users and screen sizes, and performing and implementing research for display and comparison of different fuel cycles and transition scenarios. The overall objective is to establish a powerful and flexible open source fuel cycle simulator that significantly improves communication and understanding of fuel cycle issues for both analysts and stakeholders.

IMPORTANCE OF THE SCENARIO STUDIES ON THE “BY DESIGN” PROLIFERATION RESISTANCE IMPROVEMENT

Georgios Glinatsis
ENEA, via Martiri di Monte Sole 4, IT – 40129 Bologna, Italy
Corresponding author: georgios.glinatsis@enea.it
tel. +39 051 6098641, fax +39 051 6098279

Abstract

In relatively recent times there is a growing belief that the implementation of safeguards at an early stage of the project, of a given “component” of the fuel cycle, will contribute significantly to the improvement of the proliferation resistance behaviour. At this aim, the IAEA is cooperating with the international community to develop a new process called “Safeguards by Design” to help ensure that safeguards are fully integrated into the design process of a nuclear facility from initial planning through design, construction, operation, and decommissioning. By taking into account design features that facilitate the implementation of international safeguards early in the design phase, i.e. Safeguards by Design, the proliferation resistance of the system can be improved.

Of course a complete fuel cycle should be considered in the assessment of the proliferation resistance, even if often during the nuclear reactor (= a fuel cycle component) design there is a lack of design information and specifications on fuel cycle, installations/plants and location, etc.

Moreover, from the reactor designers point of view the proliferation resistance (and physical protection), despite of their importance, are not the main technical parameters or constraints considered in the early conceptual design stage.

Because any optimisation freezes the neutron design, in order to be feasible and efficient, proliferation resistance improvement by design at core level, should be based on a clear and self-consistent set of proliferation resistance requirements (like design constraints) given at a very early stage. An approach, that could help the core designer to enhance the proliferation resistance characteristics, will be discussed. It is based on the scenario studies and information coming from these, by analysing performances and behaviour of physical “observables” parameters.

Promising and useful indications and results have been obtained by such approach application to a European level energy demand scenario.

TRANSITION FROM THERMAL TO FAST REACTORS IN FINLAND

Pauli Juutilainen
VTT Technical Research Centre of Finland

Abstract

The present study describes a basic transition path from thermal to fast reactor fleet in Finland using the fuel cycle software COSI V6 developed by CEA. The purpose of the calculation is to determine whether the domestic plutonium stockpile created in light water reactors is going to be sufficient to start fast reactors and at which time this could be possible. Another step is to estimate the maximum amount of minor actinides that could be burned in the assumed Finnish fast reactors.

The current nuclear reactor fleet in Finland consists of two VVERs with 490 MWe output each and expected lifetime of 50 years, and two 880-MWe BWRs with overall life expectancy of 60 years. All these reactors started to operate within a few-year-span around 1980. In addition to the operating reactors, a 1600-MWe EPR is under construction and estimated to start the commercial operation in 2014. Yet another two units are under consideration and thought to start the electricity production in the beginning of 2020’s. The reactor type choices between PWR and BWR are to be decided in the near future, but the power output will be around the magnitude of EPR. They are supposed to operate 60 years. The impact of the reactor type on the spent fuel composition is worth examining.

It is assumed in the present study that the planned reactors are the last LWRs to be constructed. After their retirement the energy demand is assumed to be mostly satisfied without major nuclear contribution. However, some fast reactors could be introduced at that time to incinerate the legacy waste. Some alternative scenarios including FR deployment could be based on, for example, larger contribution of nuclear power with or without LWRs beyond 2080 or foreign collaboration in waste incineration. On the other hand, a collapse of one or both of the planned new unit projects would drastically change the outcome as they are going to produce a significant fraction of the total spent fuel inventory.

ANALYSIS OF A FAST REACTOR TRANSITION SCENARIO WITH EVOLCODE/TR_EVOL

I. Merino Rodríguez, F. Álvarez-Velarde and F. Martín-Fuertes
Nuclear Innovation Program
Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas
CIEMAT. Madrid (Spain)

Abstract

A specific scenario of European Sodium Fast Reactor technology (ESFR) penetration in a European fleet has been analyzed by the Innovation Group at CIEMAT by means of two kind of computational tools, a parametric fast running computer code named ECC, under development to explore cost-benefit strategies for advanced nuclear fuel cycles, and a more accurate tool named TR-EVOL (module of the EVOLCODE 2.0 simulation system), in which reactor physics is more realistically considered.

The considered scenario assumes that there will be a European low energy-demand scenario of electricity. The same main assumptions as in the EU FP6 PATEROS project were taken into account concerning the initial condition: an association of seven European countries, together with their nuclear material stocks, is considered in 2010. Also, the electricity production is assumed to be constant during the period of reference, being 800 TWhe per year.

The main objective is to address the possibility of substitution of the light water reactor technology (LWR) by SFR over a period of 90 years (2010-2100). Also, a variation to the base case is considered for Minor Actinide (MA) management in the reactors.

Four reactor technologies are considered, including the present LWR-UOX and LWR-MOX fleets, Gen II type, both to be replaced by the European Pressurized water Reactor (EPR) design after 2020; EPR technology, Gen III+ type, each unit having 60 years operational lifetime to be replaced, in turn, in 2100 by ESFR; and the ESFR technology, assumed to be available for deployment in the year 2040. Therefore, it is introduced in that year and it should coexist with the EPR type for a number of years. In the coexistence period, the electricity contributions coming from both technologies are kept constant after the ESFR introduction phase, i.e., as of 2050: 1/3 comes from ESFR, 2/3 from EPR. After that, in 2080, the EPR technology starts to be fully replaced by the ESFR.

Both kinds of codes complement the analysis and results indicate that a successful scenario of full replacement of LWR technology by ESFR in a 90-year time frame is possible. On the one hand, the requirements for Pu breeder reactors are moderate enough for the reactor design to avoid blankets. On the other hand, the amount of Pu to be managed is very large.

When an alternative case with homogeneous transmutation strategy having a constant 2.5%w of MA content in the fuel at loading is assumed, the Np+Am stock is nearly reduced to zero after about a century (except for a small amount coming from reprocessing losses), while the Cm stock is decreased but not fully eliminated. The strategy, however, would require the fabrication of large amounts of MA-bearing subassemblies.

FUEL CYCLE STUDIES ON MINOR ACTINIDE BURNING IN GAS COOLED FAST REACTORS

Máté Szieberth, Máté Halász, Sándor Fehér, Tibor Reiss
Budapest University of Technology and Economics, Institute of Nuclear Techniques, Hungary

Abstract

The gas cooled fast reactor (GFR) is a Generation IV reactor type considered as an alternative fast neutron reactor design aimed to improve the sustainability of nuclear energy by improving the uranium utilization efficiency. Besides fuel breeding the especially fast neutron spectrum of the GFR also provides excellent opportunities for minor actinide (MA) burning. Development of the design of a 2400 MW_{th} full-scale and a 70 MW_{th} demonstrator GFR (ALLEGRO) is going on in the Euratom sponsored GoFastR project.

The paper presents fuel cycle studies performed at the Budapest University of Technology and Economics (BME) for the assessment of the transmutational capabilities of the 2400 MW_{th} GFR design in the framework of the GoFastR project. Such studies pose two main challenges:

- The evaluation of the different transmutation options can be performed based on the detailed composition of the final waste, which requires the tracking of a wide range of isotopes in the fuel cycle and the determination of the accurate composition of the spent fuel.
- Minor actinide recycling options in transmutation fuel cycles also results in a wide range of possible isotopic compositions of the core, influencing the neutron spectrum and therefore the burnup process.

Most scenario codes contain only cross-section sets at a few burnup steps, which are not flexible enough for such analysis. On the other hand detailed burnup calculations are too time consuming to be inserted in the simulation of the complete fuel cycle. In order to circumvent this problem a new approach were developed for the modelling of the reactor burnup. Multidimensional regression method was used to determine one-group cross-sections as functions of the core composition based on the results of numerous (few thousand) core transport calculations with different isotopic compositions. The set of isotopic compositions for which the core calculations were performed had to cover the space of the possible core compositions with the considered recycling options. With the help of the generated composition dependent cross-section functions, a fast and flexible burnup calculation scheme were developed which can be easily integrated into fuel cycle simulations. The developed model was verified against detailed burnup calculations with satisfactory results.

The burnup model was applied for the analysis of fuel cycle scenarios including a mixed fleet of GFRs and conventional LWRs. Homogeneous recycling of minor actinides into the GFRs was considered and their MA consumption was investigated. The results confirm the expectation that GRFs can also be applied for burning of MAs produced in other reactors. Calculations were performed with different MA ratios in the GFR core and different ration GFRs in the reactor fleet in order to find an optimum.

The present work focuses on the analysis of GFRs, but the developed methodology for burnup modelling can be applied for other reactor types, as well.

PREVENTIVE WAY OF RELATIVELY LOW RADIACTIVE WASTE ENERGY PRODUCTION BY THORIUM-URANIUM FUEL CYCLE APPLICATION

Marcin Szuta, Andrzej Wojciechowski
National Centre for Nuclear Research, Poland

Abstract

Analysis of possible ways of reduction of radioactive wastes by transmutation of radioactive long-lived fission products such as ^{99}Tc, ^{129}I and ^{135}Cs and by burning up of transuranic nuclides implies that the reactor core should consist of three zones with fast, epithermal and thermal neutron spectra. High flux thermal neutron environment ($\geq 10^{16}$ n/cm^2·s) is expected as the best way for the transmutation of most of the radioactive waste to stable or short-lived nuclides and for increasing the probability for fission such actinides as ^{237}Np and ^{238}Np.

Since we are not able to construct such a reactor core with the three zones and we are not able to reach such an intense thermal neutron flux ($\geq 10^{16}$ n/cm^2·s) in terms of technical feasibility and in view of the inefficiency of actinides incineration for a low intensity thermal neutron flux, we focus our attention on the thorium-uranium fuel cycle as a prophylactic way of energy production where the radio-toxicity of the wastes is about three orders of magnitude smaller than in the case of classical PWR reactors.

^{232}Th is a better fertile material than ^{238}U in thermal reactors because of the three times higher thermal neutron absorption cross-section of ^{232}Th (7.4 barns) as compared to ^{238}U (2.7barns). Thus, conversion of ^{232}Th to ^{233}U is more efficient than that of ^{238}U to ^{239}Pu in thermal neutron spectrum though the resonance integral of ^{232}Th is one third of that of ^{238}U. The values of cross sections in brackets refer to the thermal neutrons.

While the uranium 238-plutonium 239 fuel cycle requires fast neutrons to be sustainable, the thorium 232-uranium 233 fuel cycle is sustainable with either thermal neutrons or fast neutrons.

The ^{232}Th – ^{233}U fuel cycle let us obtain breeding of fissile atoms both in fast, epithermal and thermal neutron spectra.

As thorium based fuels have benefits in terms of radio-toxicity there are however challenges in terms of reprocessing the spent thorium based fuel. The database and experience of thorium fuels and thorium fuel cycles are very limited , as compared to UO_2 and $(U,Pu)O_2$ fuels, and need to be augmented before large investments are made for commercial utilization of thorium fuels and fuel cycles.

That is why, as the first step of studying the thorium application feasibility, once through thorium based fuel cycle analysis for energy production and radioactive waste transmutation was undertaken, which does not require the technically difficult reprocessing of the spent fuel. Justification for such a study direction is the relatively small amount of radioactive wastes during exploitation the thorium based fuel in the process of energy production and that the natural thorium is about 3.5 times more abounded than the natural uranium in the earth.

Moreover the thorium based fuel enables to increase burn-up of the fuel due to breeding of fissile atoms.

The idea is to analyze the thorium based fuel application in the open cycle feasibility study in the existing light water reactors with minimal modifications in order to exploit them.

Preliminary analysis of thorium based fuel application in the EPR reactor have shown that once through thorium fuel cycle can be reached with difficulty. It is inferred that the ^{233}U concentration tends to saturation value which does not depend on power density while the kinetics of reaching the saturation value depends on it.

Whereas the available neutron numbers for breeding in the thorium-uranium fuel cycle is only a little above zero (0.1 – 0.3, the value is not given precisely in the open literature) we have focused on the accelerator driven system (ADS) where the effect of breeding can be easier utilized in more flexible way for higher burn-up of the fuel giving impact on economy improvement. Review of available literature concerning the ADS let us include to the analysis determination of optimal spallation neutron target sizes which enables to obtain the required ion beam current of the value achievable with today’s technology.

Monte Carlo methodology calculations for the contribution to the analysis of thorium based fuel application in the EPR reactor and in the accelerated driven systems (ADS) were used.

ACTINIDE AND FISSION PRODUCT EVOLUTION BENCHMARKING WITH VANDELLOS II (PWR-SPAIN) MEASURED ISOTOPIC VALUES WITH CONSIDERING ALL THE BURN-UP HISTORY WITH CONSECUTIVE CALCULATION

Ignacio Javier Dopico Perez de Rada[1], Candan Töre
[1]SEA (Shielding Engineering and Analysis) S.L.
Avenida Atenas 75, Locales 106-107 Las Rozas-Madrid-Spain
igndopico@gmail.com candantore@yahoo.es

Abstract

High burn-up spent fuel contains a rather large amount of long-lived actinides and fission products. Prediction of these amounts is important for the purposes of transmutation, safe guards and safe storage of the spent fuel. The isotopic evolution mainly depends on the neutron spectrum which the nuclear fuel is exposed during its life in the reactor core. Prediction of the amount of the isotopes which are accumulated in the spent fuel need to be performed with validated neutronic calculation codes.

Because of the restrictions placed on and the cost associated with experimental facilities, the value of computer modelling and calculation has increased. Nowadays, it has become possible to model many varieties of nuclear systems (including full reactor core) and perform burn-up and decay calculation.

Unfortunately, it is not easy to predict perfectly isotopic contents of the irradiated fuel by calculation because of many reasons such as uncertainties of the fuel fabrication, quality and uncertainties in the basic nuclear data (cross-section, fission yields, etc.), uncertainty of the representation of the irradiation geometry, etc.

Traditionally, isotopic evolution calculation of a spent nuclear fuel is performed cycle by cycle. The calculated amount of actinides and fission products for a cycle are introduced to next modelled core cycle until the last cycle of that fuel. Through that methodology the chain of isotopic evolution is lost. Consequence of this can be a significant uncertainty is added to prediction of the amount of the actinides and fission products.

At this study, isotopic evolution of the sample E58-263 of assembly WZR0058 of Vandellos Unit II (PWR-Spain) is calculated with MONTEBURNS code system. The sample was exposed with different neutron spectrum because of its different core location at the different cycles. At the calculation, all the cycle burn-up history of the sample is considered consecutively by using the ‘remove’ and ‘add’ option of the MONTEBURNS code. The calculated results are compared with the measurements and with the cycle by cycle calculation methodology results.

Posters -
Session III

SEPARATION OF GROUP I AND II FISSION PRODUCTS FROM LiCl WASTE SALTS DELIVED ELECTROLYTIC REDUCTION PROCESS OF USED OXIDE FUEL

Y.Z. Cho, T.K. Lee, H.C. Eun, H.S. Park, I.T. Kim, K.I. Park
Korea Atomic Energy Research Institute, ROK

Abstract

During the electrolytic reduction of used oxide nuclear fuel, Group I and Group II fission products are oxidized into chlorides and partition into the molten chloride salt electrolyte, LiCl. These fission products are both highly radioactive and heat generative. Thus, LiCl containing these fission products must be disposed of in durable waste forms that are compatible with the environment inside of a geologic repository for thousands of years, which results in the formation of significant volumes of the final waste form for disposal. The purpose of this research is to develop technology for purifying the LiCl waste salt by means of layer melt crystallization process. Layer melt crystallization process no needs any chemicals or ion-exchange matrix for purifying LiCl waste salt. It just uses solubility difference of impurities between solid and melt phase. By layer melt crystallization, Group I and II FPs in LiCl waste salt can be concentrated with in small portion of LiCl waste salt, so minimization of final waste amount of LiCl waste salt can be achieved. In this present study, the effects of operating conditions such as cooling intensity, molten salt temperature and crystallization time on Group I and II fission products separation and pure LiCl salt recover rate were analyzed by using the lab-scale(3kg-LiCl/batch) layer melt crystallization apparatus.

One-Group Fission Cross Sections for Plutonium and Minor Actinides Inserted in Calculated Neutron Spectra of Fast Reactor Cooled with Lead-208 or Lead-Bismuth Eutectic

G.L. Khorasanov and A.I. Blokhin
Institute for Physics and Power Engineering (IPPE), Obninsk, Russian Federation
E-mail: khorasan@ippe.ru

Abstract

Since 1999 authors have proposed a new coolant for nuclear power installations with intermediate and fast neutron spectra [1]. This coolant is based on lead enriched with the lead isotope, lead-208, which is characterized by very low neutron capturing and slow neutron moderating. These features of lead-208 allow having some gain in the quantity of plutonium [2] to reach criticality of fast reactor (FR) or in hardening the neutron spectra of the core and its topical and lateral blankets. The gain in the neutron multiplication factor K_{ef} is about 1.5-2 percent for small sized FRs and hardening of core neutron spectra can reach about 6 percent as compared with other heavy metal coolant as natural lead or lead-bismuth eutectic.

In the present paper the one-group fission cross sections for plutonium and minor actinides inserted in calculated neutron spectra of the FR RBEC-M [3] core are given. Calculations performed show that in this type of 900 $MW_{thermal}$ reactor core the replacement of its standard lead-bismuth coolant with lead-208 leads to increasing neutron multiplication factor and neutron spectrum hardening.

Neutron spectra in both cases of coolant, Pb-208 or Pb-Bi, were calculated in using MCNP5 [4] Monte-Carlo code on the basis of the input data given in the RBEC-M project designed in the Moscow Kurchatov Institute.

The microscopic fission cross sections on the basis of files of the evaluated nuclear data for the ENDF/B-VII.0 version were taken and one-group fission cross sections in using this 28 neutron energy groups [5] were calculated.

As follows from calculations, the replacement of coolant from Pb-Bi with Pb-208 in RBEC-M leads to increasing the core mean neutron energy from its standard value 0.3992 MeV to 0.4246 MeV, i.e. on 6.4%. As concerns Pu isotopes, their one-group fission cross sections become slightly changed. More dramatically Am-241 one-group fission cross section is changed, on 9.6%, as a result of neutron spectrum hardening. It must be mentioned that 5-6 % of Am-241 is usually presented in the power grade plutonium obtained from spent fuel of PWR or VVER. Such mix of power grade plutonium and 5-6 % of Am-241 is usually proposed as a fuel for FR’s core.

Similar situation occurs in lateral blanket contained small quantities of minor actinides. From calculations performed it follows that as a result of switching coolants from Pb-Bi to Pb-208 lateral blanket mean neutron energy increases from 0.2509 to 0.2662 MeV, i.e. on 6.1%. As a result of the neutron spectrum hardening, the one-group fission cross sections of Np-237, Am-241 and Am-243 increase up to 8-11%. As is known these minor actinides need higher neutron energy, more than 0.1-0.2 MeV, to be fissile. The result of increasing MA one-group cross sections is very important as it allows reducing the time of minor

actinides burning in the FR lateral blanket for their excluding from nuclear wastes.

References

[1] G.L. Khorasanov, A.P. Ivanov, A.I. Blokhin, and A.L. Shimkevich. Lead and tin targets for reducing polonium waste. In: 3rd Int. Conf. on Accelerator Driven Transmutation Technologies and Applications. June 7-11, 1999, Praha, Czech Republic, Published by ICARIS Ltd., p.46, Paper Tu-O-E8.

[2] G.L. Khorasanov, V.V. Korobeynikov, A.P. Ivanov, A.I. Blokhin. Minimization of an initial fast reactor uranium-plutonium load by using enriched lead-208 as a coolant. Nuclear Engineering and Design, v. 239, No 9, p. 1703-1707, 2009, (ISSN 0029-5493).

[3]P.N. Alekseev, K.O. Mikityuk, A.V. Vasiljev, P.A. Fomichenko, T.D. Shchepetina, S.A. Subbotin. Optimization of Conceptual Decisions for the Lead-Bismuth Cooled Fast Reactor RBEC-M. Atomnaya Energiya, 2004, v. 97, issue 2, pp.115-125); http://www.iaea.org/NuclearPower/ SMR/crpi25001 /html/).

[4] MCNP – A General Monte Carlo N-Particle Transport Code, Version 5. X-5 Monte Carlo Team. Los Alamos National Laboratory Report LA-UR-03-1987 (April, 2003).

[5] G.N. Manturov, M.N. Nikolaev, A.M. Tsiboulia, Group constant system ABBN-93. Part 1: Nuclear constants for calculation of neutron and photon emission fields, Issues of Atomic Science and Technology, Series: Nuclear Constants, Issue 1, p. 59 (1996) (in Russian), (ISSN 0207-3668).

DECONTAMINATION OF SPENT DIAMEX SOLVENTS CONTAINING CONTAMINANTS DIFFICULT TO STRIP

Kamil Vavřinec Mareš, Jan John, Martin Vlk, Ferdinand Šebesta, Tereza Svobodová
Czech Technical University in Prague, Czech Republic

Abstract

A serious obstacle in industrialization of the DIAMEX process is accumulation of “difficult-to-strip” elements (e.g. Ru, Y, Mo, Pd, Zr, Sr etc.) in the spent solvents. The aim of this study was to verify the possibility to decontaminate such spent solvents by solid sorbents.

A thorough literature review revealed that activities in the field of separation of metals from non-aqueous media are rare. Materials, which can be used for efficient separation of metals from non-aqueous liquids, include micro- and macroporous ion exchange resins, papers impregnated with resins, and (modified) cellulose. A general observation has been that the effect of small amounts of water on the distribution equilibria can be remarkable, especially in low-polar solvents. A problem in the pre-selection of potentially suitable sorbents is the fact that for the case of spent solvents and the metals like Ru or Pd, their speciation in organic phase is hardly predictable and may well be of non-ionic nature (colloids or pseudo-colloids containing reduced species etc.).

For the experimental part of this work, the DIAMEX solvent based on TODGA in kerosene /octan-1-ol (5 vol.%) mixture was used as a reference system. An attempt to characterize speciation of the elements of interest in the simulant of the spent solvent was done by FTIR, ESI-MS, APCI-MS, and thin-layer chromatography.

More than 50 solid sorbents of various nature, identified as potentially prospective, were obtained and their efficiency for the removal of Ru and Y from the simulated spent solvent was screened in batch contact experiments. The results obtained revealed that relatively high weight distribution ratios D_g, sufficient for the design of a process for quantitative separation of the contaminants from the solvent, can be achieved for some of the solid sorbents, e.g. Amberlyst A26 or Fe-EDA-SAMMS. For the prospective materials, sorption kinetics and sorption isotherms were determined. The materials with the highest sorption capacity were tested in dynamic column experiments. The results obtained in these tests will be presented in detail.

A NEW METHOD FOR STABILIZATION/SOLIDIFICATION OF RADIOACTIVE WASTE SALT (LiCl, LiCl-KCl) FROM PYROCHEMICAL PROCESS

Hwan-Seo Park, In-Hak Cho, Su-Na Ahn, In-Tae Kim, Geun Il Park
Korea Atomic Energy Research Institute (KAERI), Korea

Abstract

A pyrochemical process to recover uranium and TRUs consists of a series of electrolytic processes using metal salts such as alkali metal chlorides. During the process, uranium and TRU elements are recovered as metallic form and a series of fission products are left in the electrolyte. This radioactive waste is one of problematic waste not to directly apply to the conventional vitrification process due to high volatility and low compatibility with silicate glass. At present, there are two approaches to the immobilization of these radioactive wastes, Cl-containing material as a host matrix and non Cl-containing matrix by dechlorination. Among many Cl-containing minerals, sodalite ($Na_8Al_6Si_6O_{24}Cl_2$) is a practical example to treat a metal chloride waste, which was suggested by Argonne National Laboratory (ANL). Also, the immobilization of metal chloride waste by using phosphate-based glass has been reported by Research Institute of Atomic Reactor (RIAR). Different from these direct immobilization, our research group adapted a dechlorination approach for waste salt by using a inorganic stabilizer, **SAP**, that composes of five kinds of oxides, SiO_2, Al_2O_3, P_2O_5, Fe_2O_3 and B_2O_3. This inorganic composite can be prepared by a conventional sol-gel process. It has a sequential bond (-Si-O-Al-O-P-) which can be broken to react with metal chloride when contacting with molten salts (LiCl or LiCl-KCl). By this reaction, a series of products (metal aluminosilicates, metal aluminophosphates and metal orthophosphates) manageable at high temperature can be obtained. A monolithic wasteform can be prepared by a heat-treatment of product at 1150°C. The wasteform composes of silicate and phosphate glassy phase uniformly distributed in tens of nm scale. Gaseous chlorine (or other halogen gas) generated during this process can captured by alkali metal oxide (Li_2O or K_2O) to produce alkali metal chloride that can be reused as an electrolyte. In conclusion, the new method suggested in this study, the dechlorination of metal chloride by using SAP and the recycle of gaseous chlorine by alkali metal oxide, can provide a chance to reduce the final volume for disposal and to maintain the total amount of Cl used in pyrochemical process.

Posters - Session IV

The use of GFR dedicated assemblies in the frame of advanced symbiotic fuel cycles: an innovative way to minimize the long-term spent fuel radiotoxicity

N. Cerullo[1], D. Chersola[2,1], G. Lomonaco[2] and R. Marotta[2,1]
[1]*DIMNP, University of Pisa, CIRTEN, Largo L. Lazzarino 2, 56126 Pisa, Italy*
[2]*DIME/TEC, University of Genova, Via all'Opera Pia 15/a, 16145 Genova, Italy*

Abstract

Am and Cm are mainly created by U and Pu transmutation. In terms of radiotoxicity, these MA are (potentially) quite dangerous for a long period. In addition, most of their isotopes are not fissile, so for their elimination only the transmutation process seems to be effective.

However while fast reactors are characterized by a high and hard neutronic flux (very good for burning Pu), Am and (above all) Cm cross sections are generally higher in a (epi-)thermal spectrum. So, in order to achieve both the targets, it is possible to introduce particular dedicated ("over-moderated") elements in a fast reactor core. The idea is to maximize the transmutation rate by using the high fluence (typical of a fast rector) in combination with a spectrum (thermal or slightly epi-thermal) optimized for burning those MA. By adopting the proposed solution it could be possible to strongly reduce Am and Cm and to produce energy at the same time.

In this respect, we found that the possibility of using dedicated assemblies (DAs) in a GFR seems to be a good solution for increasing MA elimination. The choice of positioning DAs both in central and in peripheral core zones allows a sufficient (from safety point of view) delayed neutron fraction with an adequate transmutation rate. Furthermore to load a greater MA mass in the core is preferable from the transmutation point of view.

To further improve the transmutation performance (particularly concerning the Cm^{244} elimination), the exploitation of the spontaneous decay of some MA before reprocessing the spent fuel has been studied, too.

The present research has been partially carried on in the frame of Euratom's 7th Framework Programme GoFastR project.

MINOR ACTINIDE TRANSMUTATION IN A GAS-COOLED FAST REACTOR

Zoltán Perkó and Jan Leen Kloosterman
Delft University of Technology, Department of Radiation, Radionuclides and Reactors, Mekelweg 15, Delft, Netherlands
Sándor Fehér
Budapest University of Technology, Insitute of Nuclear Techniques, Műegyetem rakpart 3, Budapest, Hungary

Abstract

The gas-cooled fast reactor (GFR) is one of the six reactor designs under investigation in the Generation IV initiative and is specifically dedicated to minor actinide (MA) transmutation and nuclear waste reduction. This paper summarizes the work done on the CEA designed GFR600 model in order to assess its MA burning capabilities using the SCALE code system.

A parametric study was performed with cores containing various amounts of minor actinides with distinct spatial distributions and of different origin. The results show that the addition of MAs to the fuel greatly reduces the reactivity loss during burnup. Moreover, the higher the MA content of the core, the higher fraction of the loaded MAs is fissioned; however, the more the safety parameters deteriorate. During single cycle irradiation only the quantity of neptunium and americium isotopes can be significantly reduced, curium isotopes accumulate. This makes reprocessing challenging as Cm isotopes are the most significant sources of both neutrons and heat in the irradiated GFR fuel.

Multiple consecutive cycles were also investigated with two refuelling strategies. Adding only depleted uranium to the reprocessed actinides (pure DU feed strategy) can reduce the total initial minor actinide content by up to 70% in the first 5 cycles of 1300 effective full power days. Moreover, the reactor can be made critical during this time if the initial MA content of the core is higher than 3%. When MAs are also added (constant MA content strategy) the reactivity has a continuous increase from cycle to cycle, primarily due to the ^{238}Pu breeding from ^{237}Np. Unfortunately the safety parameters deteriorate in both strategies during the cycles.

The effects of the origin of MAs and plutonium were also investigated. Despite the considerably different MA and Pu vectors characteristic to the spent fuel of traditional western type pressure water reactors and Russian type VVER440 reactors, no major difference was found in the overall MA transmutation. However, the Pu isotopic composition showed a strong effect on the reactivity and the delayed neutron fraction in the first cycles.

Finally, cores having non-uniform MA content were investigated. Though the MA destruction is significantly more efficient in the center of the core than at the edge due to the harder spectrum, it was found that moving some of the minor actinides from the outer regions to the inner zone only results in minor improvement in transmutation. However, the spectral changes due to the rearrangement increase the reactivity and enable reaching higher burnup and minor actinide destruction. At the same time some of the safety parameters of the reactor further degrade.

Molten Salt Fast Reactor (MSFR) based on LiF-NaF-KF

L.I. Ponomarev
National Research Center ''Kurchatov Institute", Moscow, Russia
mucatex.leonid@relcom.ru

Abstract

Recently, it was discovered that the solubility of heavy metal trifluorides (~30 mol% for PuF_3 and CeF_3, and ~40 mol% for AmF_3) in the LiF-NaF-KF eutectic salt is very high (Fig. 1). This observation offers new options for MSFR use as an element of the future nuclear power.

The initial idea of the molten salt reactor (MSR) with thermal neutron spectrum, realized as the MSRE and MSBR projects at ORNL (USA) half a century ago, has been transformed in the last decade. The several types of MSFR have been suggested, based on the carrier salts LiF, LiF-BeF_2, LiF-NaF-BeF_2, etc. with the rather high solubility of ThF_4 (~20 mol%) but with the low solubility (~1mol%) of PuF_3 and AmF_3. This prevents the realization of the U-Pu fuel cycle of MSFR and the development of the effective reactor-burner. One of possible applications of new data on the high solubility of PuF_3 and AmF_3 in LiF-NaF-KF salt obtained recently is the effective subcritical molten salt reactor-burner of transuranic elements. The fuel composition of this reactor is LiF-NaF-KF (86 mol%) – PuF_3 (7 mol%) – AmF_3 (7 mol%) and its neutron spectrum is almost fast (Fig.2). In the equilibrium regime such a reactor does not need feeding by fissile nuclides (^{239}Pu, ^{235}U, ^{233}U) and is capable of burning out about ~300 kg/GW·year of Am and Cm.

The results presented in this work are obtained by the collaboration of Russian Institutes: NRC "Kurchatov Institute" (Moscow), VNIITF (Snezhinsk, Russia), NIIAR (Dimitrovgrad, Russia), INR RAS (Troitsk), IATE (Obninsk), VNIIKHT (Moscow).

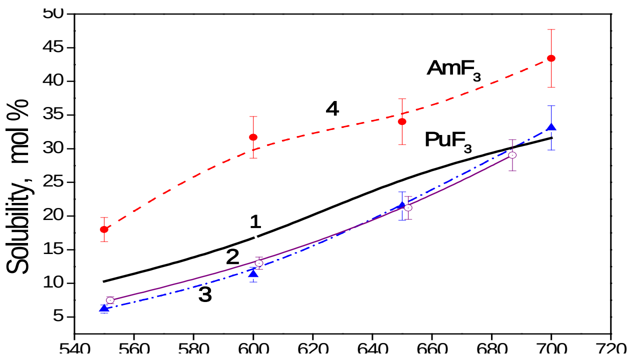


Fig.1. The calculated and measured solubility of PuF_3 and AmF_3: 1 – PuF_3 (theory, Karlsruhe), 2 – PuF_3 (NIIAR), 3 – PuF_3 (VNIITF), 4 – AmF_3 (NIIAR).

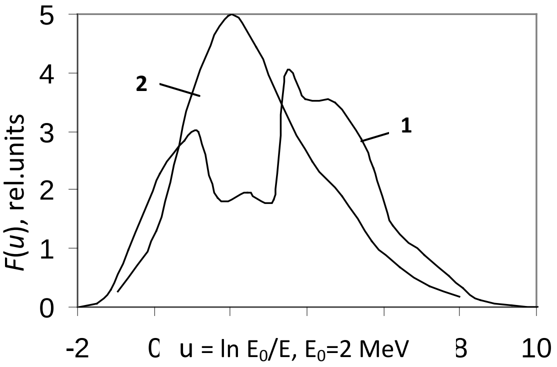


Fig.2. The neutron spectrum of the MSFR (1) in comparison with the FR spectrum (2) as a function of lethargy.

Posters -
Session V

Investigation of metal ion extraction and aggregate formation combining acidic and neutral organophosphorous reagents

A. D. Braatz*, T. Anderson, R. Ellis, M. Antonio and M. Nilsson
*Department of Chemical Engineering and Materials Science, University of California, Irvine

Abstract

Advanced nuclear fuel cycles are dependent on successful chemical separation of the various elements present in the used fuel. Numerous extraction systems have been developed for the recovery and separation of the various metal ions present in the used fuel. In these solvent extraction systems for the purification of spent nuclear material, aggregation phenomena can interfere with the efficiencies of the processes and decrease the successfulness of the separation. The predictability of these phenomena have been challenging due to the lack of a fundamental understanding of the mechanism that drives these aggregate formations. To investigate the aggregation more closely, tri-n-butyl phosphate (TBP) and dibutyl phosphonic acid (HDBP) in n-dodecane were chosen as the organic phase to be contacted with an aqueous phase containing a single metal ion from the lanthanide series in either 2M or 0.2M nitric acid. The extraction of lanthanum and dysprosium from nitrate solutions at various metal ion concentrations between 10^{-4} M and 10^{-2} M was investigated. Third phase was observed in dysprosium samples of high metal ion concentration though only slight clouding was observed in lanthanum solutions at the same concentrations and conditions. Distribution ratios of metal ions, water uptake, nitrate concentration, and acid concentration were measured to help determine physicochemical properties of the investigated system. Possible explanations of effects and future direction of this project will be presented. Separate studies of the X-ray scattering behavior of these solutions will also be presented.

AQUEOUS COMPLEXATION AND INTERACTIONS OF ND(III) AND AM(III) WITH CITRATE IN PERCHLORATE MEDIA

M. Alex Brown,[a] Alena Paulenova,[b] Artem V. Gelis[c]
[a] Department of Chemistry, Oregon State University, Corvallis, Oregon 97330, United States
[b] Department of Nuclear Engineering and Radiation Health Physics, Oregon State University, Corvallis, Oregon 97330, United States
[c] Chemical Sciences and Engineering Division, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Illinois 60439, United States

Abstract

Carboxylic acids have played an important role in the field of separating actinides (An) from lanthanides (Ln) using liquid-liquid extractions to develop effective and economical means to reducing the volume, toxicity, and lifetime of irradiated nuclear fuel in the United States.[1, 2] Studies have now shown that negligent control of the carboxylate concentration in advanced liquid-liquid reprocessing of high-level waste can jeopardize the solubility of Ln and An.[3] Recent bench-scale experiments have demonstrated that the more soluble 3-carboxy-3-hydroxypentanedioic acid (citric acid) is a propitious aqueous complexant that can effectively aid in the separation of transition metals from f-elements mixtures using a combined solvent mixture of di(2-ethylhexyl) phosphoric acid (HDEHP) and octyl(phenyl) - N′,N′,- diisobutylcarbamoylmethyl phosphine oxide (CMPO).[4] The complexation of Ln with anions of citric acid has been previously studied with conflicting results regarding the coordination of metal ions between carboxylic groups, the feasibility of protonated metal complexes, and the formation constants themselves.[5]

Using potentiometric and spectrophotometric measurements as well as specific ion interaction modeling (SIT) we report protonation equilibria of citric acid and its complexes with Nd(III) and ^{243}Am(III) using least-squares fitting of the experimental data. For trivalent metal (M^{3+}) complexation, the best model included mononuclear and protonated species

$$\mathrm{M}^{3+} + i\mathrm{H}^{+} + j\mathrm{Cit}^{3-} \rightleftharpoons \mathrm{MH}_i\mathrm{Cit}_j^{3+i\text{-}3j} \quad (1)$$

The calculated formation constants showed enhanced stability relative to other carboxylic complexes with Ln elements which was attributed to some coordination of the α-hydroxyl group of citric acid to the metal ion. Protonated metal complexes demonstrated an overall lower stability than that of the neutral charged $MCit^0(aq)$ species. A comparison of the Nd(III) and Am(III) formation constants with citrate showed very little discrepancy which is consistent given their similar charge, ionic radii, and affinity for hard oxygen-donor ligands.

References:

[1] Del Cul, G.D., Bond, W.D., Toth, L.M., Davis, G.D., Dai, S., Metcalf, D.H. Citrate based “Talspeak” Lanthanide-Actinide Separation Process. Separation Science & Technology 1997, 32, 431-446.

[2] Weaver, B., Kappelmann, F.A. Talspeak: A New method of Separating Americium and Curium from the Lanthanides by Extraction from an Aqueous

Solution of an Aminopolyacetic Acid Complex with a MonoAcidic Organophosphate or Phosphonate. Oak Ridge National Laboratory, 1964, 3559.

[3] Arai, K, Yamashita, M., Hatta, M., Tomiyasu, H., Ikeda, Y. Modified TRUEX Process for the Treatment of High-Level Liquid Waste. Journal of Nuclear Science & Technology 1997, 34, 521-526.

[4] Brown, M.A., Gelis, A.V., Paulenova, A.P., Vandegrift, G.F. Combining TRUEX and TALSPEAK in One Process. 16th Symposium on Separation Science and Technology for Energy Applications, Gatlinburg, TN. 2009

[5] Wood, S. The Aqueous Geochemistry of the Rare Earth Elements: Critical Stability constants for Complexes with Simple Carboxylic Acids at 25C and 1 bar and their Application to Nuclear Waste Management, Engineering Geology 1993, 34, 229-259.

A *N,N´*-di-*n*-OKTYL-TODGA-dicarbollide Extractant for Actinide(III) Separation from Acidic Waste Solutions

Mária Bubeníková[1], Pavel Selucký[1], Jiří Rais[1], Bohumír Grüner[2], Petr Švec[2]
[1]Nuclear Research Institute Řež plc., Czech Republic
[2]Institute of Inorganic Chemistry, Academy of Sciences of the Czech Republic, v.v.i., Czech Republic

Abstract

Chemical partitioning of minor actinides from other fission products, in particular from lanthanides, is many-faceted challenge for existing nuclear fuel treatments. In recent years, several HLLW partitioning processes such as TRUEX, TRPO, DIDPA, DIAMEX, TODGA/ TBP utilizing variety of extractants have been developed for actinide separations. However, none of them provided any substantial actinide (III)/ lanthanide (III) selectivity. Processes such as SANEX or ALINA have been proposed for subsequent selective separation of actinides(III) from lanthanides(III).

Recently, the extractants based on two cobalt bis(dicarbollide) (1^-) ions covalently bound to organic, diglycolyl acid platform (with different alkyl/ aryl substituents on the amide nitrogen) by amidic bonds, have been developed for use in nuclear treatment. In this work, the extractant *N,N´*-di-*n*-oktyl-TODGA-COSAN of the formula $\{[(N,N'\text{-}(8\text{-}(OCH_2CH_2)_2\text{-}1,2\text{-}C_2B_9H_{10})(1',2'\text{-}C_2B_9H_{11})\text{-}3,3'\text{-}Co)(N,N'\text{-}n\text{-}C_8H_{17})NCOCH_2]_2O\}Na_2$ was evaluated for actinides(III) separation from acidic aqueous waste solutions. The extractant provided sufficient solubility in an ecological low polar solvent mixture of hexyl methyl ketone and *n*-dodecane and exhibited good hydrolytical stability. The *N,N´*-di-*n*-oktyl-TODGA-COSAN enabled good separation of actinides(III) and lanthanides(III) from bulk of the fission products present in the model PUREX feed. The co-extraction of some undesirable elements as Zr, Mo, Pd was suppressed by using oxalic acid and HEDTA in the feed solution. The *N,N´*-di-*n*-oktyl-TODGA-COSAN, similarly as other TODGA-like compounds, did not show any substantial actinide(III)/ lanthanide(III) selectivity. Therefore, for selective separations of actinides(III) from lanthanides(III) the hydrophilic complexing agent of diethylene-triamine-N,N,N´,N´´,N´´-pentaacetic acid (DTPA) with sodium nitrate alone or in combination with different carboxylic buffers have been tested. It was found that Am(III) could be selectively stripped from loaded organic phase by using the solution of 0.05 M DTPA buffered with 1 M citric acid and 1 M $NaNO_3$ at pH 3.0 ($SF_{Eu/Am}$ = 13). A high sensitivity of trivalent metal extraction to pH changes at the stripping step might be mentioned here as a possible drawback of the system.

A kinetic study on the extraction of Americium(III) into CyMe$_4$-BTBP

Andreas Geist, Daniel Magnusson, Udo Müllich
Karlsruhe Institute of Technology, INE, P.O. Box 3640, 76021 Karlsruhe, Germany

Abstract

CyMe$_4$-BTBP is the current European reference molecule used in the development of *r*-SANEX processes for separating Am(III) and Cm(III) from Ln(III). Successful spiked and hot tests were performed at Jülich [1] and ITU [2]. Also, a successful spiked 1c-SANEX process, directly extracting only Am(III) and Cm(III) from a simulated PUREX raffinate, was performed at Jülich [3]. However, due to CyMe$_4$-BTBP's rather slow extraction kinetics [4], comparatively low flow rates had to be used. Quantitative kinetic investigations on the extraction of Eu(III) into CyMe$_4$-BTBP in a rotating membrane cell support this [5].

Since no quantitative kinetic data on Am(III) were available, we studied extraction and stripping kinetics for the system Am(III) – NO_3^- / CyMe$_4$-BTBP – TODGA – diluent in the INE stirred cell [6]. Such kinetic data are important to perform highly reliable flow-sheet calculations. Extraction rates from 1 mo/L HNO_3 were measured as a function of nitrate concentration (via addition of NH_4NO_3), CyMe$_4$-BTBP concentration, TODGA concentration, and diluent (standard diluent was 1-octanol; cyclohexanone was also tested). Am(III) stripping rates into dilute HNO_3 (from a loaded phase with or without TODGA) or into glycolate solution were also measured. The following results were obtained:

- With an organic phase consisting of CyMe$_4$-BTBP in 1-octanol, Am(III) extraction rate increase first order both with *[CyMe$_4$-BTBP]* and with the interfacial area. Nitrate concentration has a small effect.
- With an organic phase consisting of CyMe$_4$-BTBP + TODGA in 1-octanol, Am(III) extraction rate is practically independent of *[CyMe$_4$-BTBP]*. However, Am(III) extraction rate increases first order both with *[TODGA]* and with the interfacial area. TODGA has a positive kinetic effect; e.g., Am(III) extraction rate increases by approx. one order of magnitude when 5 mmol/L TODGA is added to the organic phase.
- Replacing the diluent 1-octanol with cyclohexanone has a positive kinetic effect; extraction rate is slightly higher than that for an organic phase containing 10 mmol/L TODGA in 1-octanol.
- TODGA also accelerates stripping into dilute HNO_3 by approx. an order of magnitude. However, stripping into 0.5 mol/L glycolate solution (pH = 4) results in an even higher stripping rate.

All extraction and stripping rates (except for stripping into glycolate solution) are below 10^{-6} m/s. This indicates chemical regime. However, the stripping rate into glycolate solution is $2.5 \cdot 10^{-6}$ m/s, indicating a significant diffusive influence [**Error! Bookmark not defined.**].

References:

1 G. Modolo, M. Sypula, A. Geist, C. Hill, C. Sorel, R. Malmbeck, D. Magnusson, M.R.S. Foreman, *Development and demonstration of a new SANEX process for actinide(III)/lanthanide(III) separation using a mixture of* $CyMe_4$*-BTBP and TODGA as a selective extractant.* In: *Actinide and Fission Product Partitioning and Transmutation, 10th Information Exchange Meeting,* Mito, Japan, 6–10 October 2009. NEA No. 6420, OECD-NEA (2010), 235–241.

2 D. Magnusson, B. Christiansen, M.R.S. Foreman, A. Geist, J.-P. Glatz, R. Malmbeck, G. Modolo, D. Serrano-Purroy, C. Sorel, *Demonstration of a SANEX process in centrifugal contactors using the* $CyMe_4$*-BTBP molecule on a genuine fuel solution.* Solvent Extr. Ion Exch. **2009,** 27 (2), 97–106.

3 A. Wilden, M. Sypula, G. Modolo, A. Geist, *Direct actinide(III) separation from PUREX raffinate using a BTBP/TODGA solvent.* Proc. 19th Internat. Solvent Extraction Conf. (ISEC 2011), Santiago, Chile, 3–7 October 2011.

4 A. Geist, C. Hill, G. Modolo, M.R.S. Foreman, M. Weigl, K. Gompper, M.J. Hudson, C. Madic, *6,6′-Bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo-[1,2,4]triazin-3-yl)[2,2′]bipyridine, an effective extracting agent for the separation of americium(III) and curium(III) from the lanthanides.* Solvent Extr. Ion Exch. **2006,** *24* (4), 463–483.

5 F. W. Lewis, L. M. Harwood, M. J. Hudson, M. G. B. Drew, J. F. Desreux, G.Vidick, N. Bouslimani, G. Modolo, A. Wilden, M. Sypula, T.-H. Vu, J.-P. Simonin, *Highly Efficient Separation of Actinides from Lanthanides by a Phenanthroline-Derived Bis-Triazine Ligand.* J. Am. Chem. Soc. **2011,** *133,* 13093–13102.

6 M. Weigl, A. Geist, U. Müllich, K. Gompper, *Kinetics of americium(III) extraction and back extraction with BTP.* Solvent Extr. Ion Exch. **2006,** *24* (6), 845–860.

EXPLORATION OF A NOVEL SOFT DONOR EXTRACTANT FOR PRECIOUS METAL RECOVERY IN SPENT NUCLEAR FUEL AND ELECTRONIC WASTE REPROCESSING

Brian Gullekson[a], Alena Paulenova[a], P. Zalupski[b]
[a]Oregon State University, Corvallis, OR, USA
[b]Idaho National Laboratory, Idaho Falls, ID, USA

Abstract

Spent nuclear fuel reprocessing streams and electronic waste present a viable, and potentially more abundant than in nature, source of precious metals [1]. Solvent extraction with soft electron donor ligands has been shown to preferentially extract softer Lewis acid metals over harder electron acceptors as dictated by HSAB theory [2]. Thiolating oxygen sites on organic ligands is an effective method of creating softer electron donor ligands due to the lower electronegativity of sulfur. Several studies have been performed that show that thiolated ligands either more preferentially extract trivalent actinides over lanthanides, or can be used to isolate precious metals (Ag, Au, etc.) from mixed waste streams.[3],[4]

In this study, a novel ligand, bis-(dibutanethiolthiophosphonato)methane (bDBTPM)was synthesized in an attempt to recover noble metals from both spent nuclear fuel and electronic waste. The ligand is synthesized in a two step reaction, first reacting butyl mercaptan with a chlorinated diphosphineto create a thiophosphonite, followed by a second thiolating reaction to create a thiophosphonate. This reaction was confirmed by both ^{1}H and ^{31}P NMR spectrometry. Following this synthesis, lab scale solvent extraction experiments were performed with several precious metals in aqueous nitric acid solutions to determine their extractability into an organic phase of bDBTPM in n-dodecane. Preliminary results indicate that this ligand is effective at extracting precious metals at lower acid concentrations, possibly indicating its use as an extractant for use in post-PUREX waste reprocessing streams or in a solvent extraction scheme to reprocess electronic components dissolved in an aqueous phase.

References

[1] Belyaev, A. V. “Technological Problems of Platinum Metals in Nuclear Fuel Waste Disposal,” *Journal of Structural Chemistry*, Vol. 44, No. 1, 29-36, 2003.

[2] Atkins, P.; et. al. *Inorganic Chemistry, 4th Ed.*New York, W. H. Freeman & Co. 2006.

[3] Ionova, G.; et. al., “Mechanism of Trivalent Actinide/Lanthanide Separation using bis(2,2,4-trimethylpentyl)dithiophosphinic acid (Cyanex 301) and Neutral O-Bearing Co-Extractant Synergistic Mixtures,” *Solvent Extraction and Ion Exchange*, Vol. 19, Iss. 3, 391-414, 2001.

[4] Fleitlikh, I. Y.; et. al. “Cobalt and Nickel Recovery from Sulfate Media Containing Calcium, Manganese, and Magnesium with a Mixture of CYANEX 301 and a Trialkylamine,” *Solvent Extraction and Ion Exchange*, Vol. 29, Iss. 5/6, 782-799, 2011

Removal of Uranium from Thorium-Uranium SIMFUEL by Fluoride Volatility

Holly Hamilton,
Atomic Energy of Canada Limited

Abstract

One of the most significant impediments to the implementation of a sustainable thorium-oxide based fuel cycle is the lack of an industrially viable discharged fuel recycling process. The major difficulty lies in the aggressive nature of the reagents required for the initial fuel dissolution step. The concentrated nitric acid containing a small amount of hydrofluoric acid necessary to dissolve the solid fuel presents material challenges from the corrosion perspective, and process problems from the chemistry perspective. In this work a simple and effective separation of uranium from a solid solution of uranium and thorium oxide will be presented. Separations of >99 % have been achieved on thorium-uranium SIMFUEL (SIMulated-irradiated FUEL). The paper will describe the starting material, experimental apparatus, experimental conditions and results. This work is preliminary in nature but the promising results have significant implications and warrant further study and development.

A FEASIBILITY STUDY ON IODINE BEHAVIOR DURING PYROPROCESSING OF SPENT METALLIC FUEL

EUNG HO KIM
Korea Atomic Energy Research Institute

Abstract

Korea has 21 nuclear power plants in operation and as of 2011 about 11,000 ton of spent nuclear fuel was stored at four reactor sites. Currently, spent nuclear fuels are being continuously cumulated with stored at the rate of about 700 ton a year. However, it is reported that the storage pools will begin to be saturated from 2016 and thereby Korean government is on the point to decide the issues for a long-term management of spent nuclear fuel. As a promising option, Korea is developing pyroprocessing technology connected with sodium cooled fast reactor to transmute long-lived nuclides as well as minimize high-level nuclear waste amounts to be disposed of.

Among long-lived nuclides, iodine-129 has a very long half life of 15.7×10^6 years and a low cross section for transmutation as well as an impact on a biosphere for a long time even after disposing of it in a deep-geological repository. Therefore, iodine-129 needs to be managed carefully.

Information on a chemical behaviour of iodine in irradiated metallic fuel based on U-TRU-Zr alloy has a little been known, compared to that in oxide fuel. In particular, iodine chemical behaviour in a molten salt bath during pyroprocessing has been never reported and a way for isolation and recovery of iodine liberated from the molten salt was not known as well. In this respects, this study invested a various chemical behaviour of iodine not only in irradiated metallic fuel itself but also in the course of pyroprocessing in terms of thermodynamic feasibility assessment.

It is expected that iodine-129 generated as fission product will mostly exist in as metal iodides in the metallic fuel such as UI_3, ZrI_2, PuI_3, CsI, etc. Although iodine is released from metallic fuel to a gap between cladding and metallic fuel, just a little amount of iodine will be liberated into sodium bond material and produce NaI by a reaction of iodine with sodium.

As a result of thermodynamic prediction, we can anticipate that the incorporated iodine will be ultimately converted to potassium iodide (KI) in the molten salt (LiCl-KCl) in the course of electrorefining with accumulating sodium in the salt.

On the other hand, the cumulated iodine can be easily separated from molten salt. Chlorine gas can substitute iodides with chlorides and subsequently iodine is released from molten salt as off-gas. Hence, the released iodine will be easily captured and recovered by an appropriate way.

Through this procedure, iodine-129 arising from irradiated metallic fuel can be easily managed.

In addition, this study will propose a new molten salt other than LiCl-KCl eutectic salt that is generally being used in electrorefining for recovery of

metallic fuel. As substantial amounts of sodium are incorporated and increasingly cumulated in the salt during electrorefining, this can give rise to a change of various physical and chemical properties of molten salt such as melting temperature, viscosity, density, etc.

SIMULATOR OF THE BEHAVIOR OF CURIUM COMPOUNDS IN NaCl-2CsCl EUTECTIC

M.V. Kormilitsyn, Ya.M. Luk’yanova[1], A.G. Osipenko[1], A.A. Mayershin[1], V.V. Smolenski[2], A.V. Novoselova[2], [1]
[1] JSC “State Scientific Center Research Institute of Atomic Reactors”, Dimitrovgrad, 433510, Russia
[2] Institute of High-Temperature Electrochemistry UD RAS, Russia, 620219, Ekaterinburg,

Abstract

Curium isotopes in nuclear spent fuel have a large heat generation and long half-life, so that’s why it is necessary to separate them from highly active waste and then to transmute. Thermodynamic data of the formation oxygen and free-oxygen curium compounds are absent in molten chlorides.

In this work we have studied the chemical behaviour of various curium in NaCl-2CsCl eutectic and have drawn potential-oxoacidity-temperature diagram.

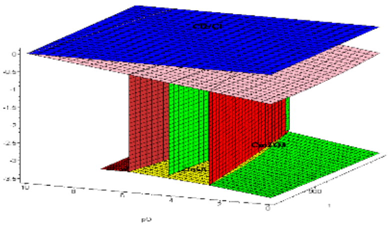
The investigations were carried out in the cell, containing two platinum-oxygen electrodes with solid electrolyte membranes. The electrodes were made from ZrO_2 membrane stabilized by Y_2O_3 (Interbil S.A.). The first electrode is used as the indicating electrode for measuring the oxygen ions activity in the investigated melt and the second – as an oxygen pump. Sometimes BaO is used as the donor of the oxygen ions.

The reactions of interaction between compounds are the following:

$Cm^{3+}_{(l)} + O^{2-}_{(l)} \Leftrightarrow CmO^{+}_{(l)}$ T = 923 K $pK_{(CmO+)} = 3.4 \pm 0.2$ (1)

$Cm^{3+}_{(l)} + O^{2-}_{(l)} + Cl^{-}_{(l)} \Leftrightarrow CmOCl_{(s)}$ T = 923 K $pK_{(CmOCl)} = 7.5 \pm 0.2$ (2)

$2Cm^{3+}_{(l)} + 3O^{2-}_{(l)} \Leftrightarrow Cm_2O_{3(s)}$ T = 923 K $pK_{(Cm2O3)} = 18.5 \pm 0.3$ (3)



Three-dimensional equilibrium E-pO-T diagram summarized the stability areas of curium compounds in the melt at different temperatures.

Removal of Adhered Salt from Uranium Deposits in Pyroprocess

Kwon*, S.W., Park, K.M., Jeong, J.H., Lee, H.S., and Kim ,J.G.
Korea Atomic Energy Research Institute, Yuseong, Daejeon, 305-353, Korea
swkwon@kaeri.re.kr

Abstract

Pyroprocessing has been developed for the recovery of actinide elements from spent fuel due to its advantages of a compactness, a nuclear proliferation resistance, and a reduction of a secondary waste generation [1-2]. It was proposed to increase the throughput of the salt removal process by the separation of the liquid salt prior to the distillation of the LiCl-KCl eutectic salt from the uranium deposits in this study. The feasibility of liquid salt separation was examined by salt separation experiments on a stainless steel sieve. It was found that the amount of salt to be distilled could be reduced by the liquid salt separation prior to the salt distillation. It was found that the liquid salt can be separated from the uranium dendrites above 500℃. The residual salt remained in the uranium deposits after the liquid salt separation was successfully removed by the vacuum distillation. It was concluded that the combination of a liquid salt separation and a vacuum distillation process is an effective route for the achievement of a high throughput performance in the salt separation process because the amount of salt to be distilled in the uranium deposits can be highly reduced and the burden of salt distiller can be reduced.

REFERENCES

[1] S. W. Kwon, E. H. Kim, and H. G. Ahn (2009), *J. Ind. Eng. Chem.*, Vol 15, 86-91

[2] B. R. Westphal, D. Vaden, T. Q. Hua, J. L. Willit, and D. V. Laug (2002), “Recent development at the cathode processor for spent fuel treatment”, *American Nuclear Society Fifth Topical Meeting,* Charleston, South Carolina, USA, Sep. 17-20

Development of an integrated sieve-crucible assembly for the sequential operation of a liquid salt separation and a distillation

S. W. Kwon*, K. M. Park, J. H. Jeong, H. S. Lee, and J. G. Kim
Korea Atomic Energy Research Institute, Yuseong, Daejeon, 305-353, Korea
swkwon@kaeri.re.kr

Abstract

The solid cathode processing is necessary to separate the salt from the cathode for the preparation of the uranium ingot with a high purity. It is indispensable to increase a throughput of the salt removal process from uranium deposits which is generated on the solid cathode of electro-refiner in pyroprocessing [1-2]. In this study, an assembly composing a liquid separation sieve and a distillation crucible was developed for the sequential operation of a liquid salt separation and a vacuum distillation in the same tower. The feasibility of the assembly was examined by the rotation test and sequential operation of a liquid salt separation and a vacuum distillation in the distillation tower. The assembly rotated successfully with the aid of the wire connected to the pulling bar placed in the exterior of the tower without a degradation of the vacuum level by the leak. The seal was assured by double O-rings during the movement of the wire through the wall of the flange. The experiment on the sequential operation of the liquid salt separation and salt distillation was carried out using an integrated sieve-crucible assembly. The uranium deposits are placed into the sieve side of the assembly and the adhered salt is separated by heating on the sieve. Then, the uranium deposits are moved to the crucible by the rotation of the sieve-crucible assembly and the residual salt is evaporated at an elevated temperature. The remained salt in the uranium deposits was further separated further by evaporation in the distillation tower for two hours at 850 °C. The adhered salt in the uranium deposits was removed successfully. The salt content in the deposits was below 0.1 wt% after the sequential operation of the liquid salt separation and salt distillation. This residual salt after salt separation process can be removed completely during the melting of uranium metal in the following ingot preparation process. From the results of this study, it could be concluded that the efficient salt separation work could be realized by the sequential operation of liquid salt separation and vacuum distillation in one distillation tower because the operation procedure was simplified and the step of cooling and reheating was not necessary for the change of the operation mode.

REFERENCES

[1] S. W. Kwon, E. H. Kim, and H. G. Ahn (2009), *J. Ind. Eng. Chem.*, Vol **15**, 86-91

[2] S. W. Kwon, J. Y. Kim, H. S. Lee, D. Ahn and H. G. Ahn (2010), *J. Radioanal. Nucl. Chem.*, Vol **284**, 143-147

IDENTIFICATION OF N,N`-DIETHYL-N,N`-DITOYLDIPICOLINAMIDE DEGRADATION PRODUCTS BY GAS CHROMATOGRAPHY-MASS SPECTROSCOPY

Joseph Lapka[a], **Alena Paulenova**[b]
a) Department of Chemistry, Oregon State University, Corvallis, Oregon 97330, United States
b) Department of Nuclear Engineering and Radiation Health Physics, Oregon State University, Corvallis, Oregon 97330, United States

Abstract

Present research has led to the development of new N-containing reagents and methods with significant potential for accomplishing separation of trivalent metals from wastes such as substituted malonic diamides and tetra-alkyl-diglycolamides (TODGA). Substituted diamides of dipicolinic acid are of interest due to their pyridine nitrogen in proximity to the carbonyl allowing it to possibly participate in coordination.

Three diamidic derivative isomers of dipicolinic acid, N,N`-diethyl-N,N`-ditoyldipicolinamide (EtTDPA) were subjected to gamma ray radiolysis and acidic hydrolysis by nitric acid in the presence of the solvent trifluoromethylphenyl sulfone (FS-13). Samples were then analyzed by GC-MS in order to determine specific degradation products in both cases. No significant differences were observed between radiolysis products of the three isomers. At doses of 200 kGy the primary radiolysis products are formed by cleavage of the carbon-nitrogen amidic linkage in both acid contacted and non-contacted samples, forming the corresponding carboxylic acid and amine groups. Other observed products are presented and discussed. The hydrolysis pathway products also show mainly the production of the carboxylic acid and amine groups by well-known amidic hydrolysis mechanisms.

LOADING AND FLOW-RATE DEPENDENCE OF THE TRANSFER RATES IN A CENTRIFUGAL CONTACTOR USING ORGANIC LIGAND CYME4-BTBP

Daniel Magnusson[1], Andreas Geist[1], Rikard Malmbeck[2]
[1]Karlsruher Institut für Technologie, Institut für Nukleare Entsorgung, 76021 Karlsruhe, Germany
[2]European Commission, Joint Research Centre, Institute for Transuranium Elements, 76344 Karlsruhe, Germany

Abstract

Centrifugal contactors are efficient and compact devices for running hydrometallurgical separation processes. Due to the short residence time in the centrifugal contactors the equilibrium distribution ratios are usually not reached. This can result in huge errors in the flow-sheet calculations which lead to badly designed processes. An estimation of the apparent distribution ratios can however be done by using stage efficiencies. The stage efficiencies depend on many different parameters and among those are the loading and the flow-rates.

$CyMe_4$-BTBP is the current European reference extracting agent for the development of trivalent actinide separation processes such as r-SANEX [1],[2] and 1c-SANEX [3]. The $CyMe_4$-BTBP ligand is known to have slow transfer kinetics and therefore has this system's kinetics been investigated further. Experiments were run in a single stage centrifugal contactor to see the influence of different metal loadings of the ligand, different flow-rates and different flow-rate ratios. The results indicate that the stage efficiency is strongly dependent on flow-rates and is also decreased with higher metal loading.

References

[1] G. Modolo, M. Sypula, A. Geist, C. Hill, C. Sorel, R. Malmbeck, D. Magnusson, M.R.S. Foreman, *Development and demonstration of a new SANEX process for actinide(III)/lanthanide(III) separation using a mixture of $CyMe_4$-BTBP and TODGA as a selective extractant.* In: *Actinide and Fission Product Partitioning and Transmutation, 10th Information Exchange Meeting,* Mito, Japan, 6–10 October 2009. NEA No. 6420, OECD-NEA (2010), 235–241.

[2] D. Magnusson, B. Christiansen, M.R.S. Foreman, A. Geist, J.-P. Glatz, R. Malmbeck, G. Modolo, D. Serrano-Purroy, C. Sorel, *Demonstration of a SANEX process in centrifugal contactors using the $CyMe_4$-BTBP molecule on a genuine fuel solution.* Solvent Extr. Ion Exch. **2009,** 27 (2), 97–106.

[3] A. Wilden, M. Sypula, G. Modolo, A. Geist, *Direct actinide(III) separation from PUREX raffinate using a BTBP/TODGA solvent.* Proc. 19th Internat. Solvent Extraction Conf. (ISEC 2011), Santiago, Chile, 3–7 October 2011.

TECHNOLOGICAL VERIFICATION OF FLUORIDE VOLATILITY METHOD

Martin Mareček, Jan Škarohlíd
Nuclear Research Institute Rez plc, Czech Republic

Abstract

Fluoride Volatility Method is considered to be a prospective advanced reprocessing technology for spent nuclear fuels of FBR systems. The experimental verification of the Fluoride Volatility Method has played an important role in Czech R&D devoted to Partitioning and Transmutation. The goal of the experimental technological line FERDA (Fluoride Experimental Research and Development Assembly), constructed in Nuclear Research Institute Rez, is verification of technology of FVM for the requirements of pyrochemical partitioning. The technology is based on the flame fluorination of spent fuel, subsequent condensation and multistage distillation of formed volatile fluorides. The powdered fuel is fed into the flame fluorination reactor together with fluorine gas. A separation of volatile fluorides from non-volatile ones carries out in this reactor. A main goal of the partitioning of volatile and non-volatile products of fluorination is to separate uranium in the form of volatile UF_6 from plutonium, minor actinides and most of fission products that form predominantly non-volatile fluorides. UF_6 is then purified from rest of volatile flurides by distillation process. The short-term capacity of the fluorination reactor is 1 – 3 kg of spent fuel per hour, however the whole facility is constructed as a batch process. The paper describes the experience with design and construction of the FERDA facility and evaluates the results of the first experiments with simulated fuel. Proposal of detailed experimental programme for the next years is described as well.

DETERMINING THE THERMODYNAMICS OF EXTRACTION OF AMERICIUM AND CURIUM BY HDEHP FROM NITRATE SOLUTION

Leigh R. Martin, Travis S. Grimes and Peter R. Zalupski
Idaho National Laboratory, Aqueous Separations and Radiochemistry Department,
Idaho Falls, ID, USA

Abstract

With the energy difference associated with a successful metal ion separation in a solvent extraction process being relatively small, paying careful attention to the thermodynamic properties of such systems ($\Delta G°$, $\Delta H°$ and $\Delta S°$) assists in providing an insight into the driving forces behind the partitioning process. While utilizing van't Hoff analysis from radiotracer studies give a reasonable approximation of how the metal ion will behave in a separations system, they do not give absolute values. Those types of experiments require a direct measurement of the reaction under study. Americium and curium are two of the key actinides that are targeted for separation in the TALSPEAK process, but the chemical interactions of these elements with the ligands that facilitate this process (lactic acid, DTPA, and di-2- ethyl hexyl phosphoric acid HDEHP) are not well defined. To that end, the study on the direct measurement of the heat of extraction of americium and curium by HDEHP has been highlighted as an area that could expand this field of research. By studying the thermochemistry of Am and Cm extraction from nitrate media by HDEHP using a calorimetric approach and van't Hoff analysis, we have begun to further expand our thermodynamic understanding of this liquid-liquid extraction system. The results from the two methodologies used for the determination of the ΔH extraction will be compared and the challenges of thermochemical evaluation of liquid-liquid distributions will be summarized.

SELECTIVE UPTAKE OF CESIUM BY HYBRID ADSORBENTS ENCLOSING HETEROPOLYACID

Hitoshi Mimura, Shun Kanome, Yuichi Niibori
Graduate School of Engineering, Tohoku University, Japan

Abstract

The selective separation and recovery of heat-generating nuclide (^{137}Cs) from high-level liquid waste (HLLW) containing highly concentrated HNO_3 and $NaNO_3$ are vital issues in relation to the partitioning of radionuclides. Ammonium molybdophosphate ($(NH_4)_3PO_4 \cdot 12MoO_3$, AMP), a fine crystal of heteropolyacid, having a high selectivity toward Cs^+ ions, is one of the most promising adsorbents for this purpose. In order to granulate the fine AMP crystals, they can be incorporated into the macrospores of zeolite (mordenite) by sol-gel method using alginate gel polymer matrices. The present paper deals with the novel preparation methods for the hybrid adsorbents enclosing AMP fine crystals, their stability and the selective uptake properties for Cs^+.

The mordenite enclosing AMP (AMP-M), a kind of inorganic/organic composites, was easily prepared by the successive impregnation of 1) PMA ($H_3Mo_{12}O_{40}P$) and 2) kneaded sol (NH_4NO_3 and sodium alginate (NaALG)) into the mordenite macropores. In this procedure, both synthesis and loading of AMP into the macropores are simultaneously accomplished. The synthetic reaction of AMP could be expressed as $H_3Mo_{12}O_{40}P + 3NH_4NO_3 \leftrightarrow (NH_4)_3PO_4 \cdot 12MoO_3 + 3HNO_3$, and the XRD intensities of AMP were almost constant up to third loading run. Here the AMP-M composites obtained by 1st to 3rd cycles were abbreviated as AMP-M-1, -2 and -3, respectively. AMP-M composites were stable up to 200°C and had high uptake ability for Cs^+ up to 2.1×10^7 R irradiation (^{60}Co).

The uptake of Cs^+ for AMP-M from HNO_3 and $NaNO_3$ solutions was examined by the batch method. The uptake of Cs^+ for AMP-M in the presence of 0.01~5 M HNO_3 attained equilibrium within 5 h, and a relatively large uptake percentage above 90% ($K_d > 10^3$ cm^3/g) was obtained; the uptake rate and uptake (%) of Cs^+ for AMP-M were markedly enhanced compared to those for mordenite matrices. The AMP-M composites also exhibited relatively large K_d values above 10^2 cm^3/g for Cs^+ even in the presence of 0.01~3 M $NaNO_3$. The uptake ability of different metal ions for AMP-M was compared in the presence of 0.01~5 M HNO_3. The order of K_d value was $Cs^+ >> Rb^+ > Ag^+$; significant difference in K_d value between Cs^+ and other cations was observed. The maximum separation factors of Cs^+/Rb^+ ($K_{d,Cs}/K_{d,Rb}$) and Cs^+/Ag^+($K_{d,Cs}/K_{d,Ag}$) were estimated to be 23.9 and 100, respectively. The selective uptake (%) of Cs^+ from simulated HLLW (SW-11E, 28 components, 2 M HNO_3, 1 M $NaNO_3$, JAEA) was estimated to be 70~80% for AMP-M-1~3.

For the practical use, the column adsorption properties (breakthrough and elution) were examined in the presence of 2.5 M HNO_3. The breakthrough and total capacities were estimated to be 0.081 and 0.22 mmol/g from the symmetrical S–shaped curves, respectively. Further, the adsorbed Cs^+ on the AMP-M-3 column can be eluted by passing the eluent of 5 M NH_4NO_3 solution and the elution (%) was estimated to be 95.4%.

Thus, the AMP-M composites seem to be effective for the selective separation of Cs^+ ions from HLLW.

ELECTROCHEMICAL AND THERMODYNAMIC PROPERTIES OF NEODIMIUM IN MOLTEN CHLORIDES

A. Novoselova, V. Smolenski
Institute of High-Temperature Electrochemistry UD RAS, Russia

Abstract

Pyrochemical separation processes in molten salts, particularly molten chlorides, have more recently been proposed as a promising option in the nuclear fuel cycle for the future, mainly due to progress in the assessment of new concepts for transmutation and the corresponding fuel cycles and several processes have already been developed for the recovery of minor actinides from spent nuclear fuels and high level radioactive liquid wastes. Nd is one of major fission product elements having larger neutron capture cross sections, and hence its removal in the pyrochemical reprocessing is desired.

The goal of these investigations is to determine the electrochemical properties of neodymium (III) and the stability of neodymium (II) ions in fused CsCl and LiCl-KCl-CsCl eutectic at large temperature range by transient electrochemical technique and *emf* method.

It was established by transient electrochemical technique that on an inert electrode Nd^{3+} ions are reduced to metallic neodymium through two consecutive steps:

$$Nd^{3+} + \bar{e} \rightarrow Nd^{2+} \qquad (1)$$

$$Nd^{2+} + 2\bar{e} \rightarrow Nd^{0}_{(metal)} \quad .. \quad (2)$$

The following disproportionate reaction of Nd^{2+} takes place in molten salts at high temperatures:

$$3\,NdCl_{2(l)} = 2\,NdCl_{3(l)} + Nd^{0}_{(metal)} \quad ...... \quad (3)$$

It has been shown that the reaction (3) is shift to the right and completed within several minutes in both solvents. So the compound $NdCl_2$ is not stable in investigated melts at temperatures above 798 K.

The value of an apparent standard redox potential of $E^{*}_{Nd^{3+}/Nd^{2+}}$ *vs.* Cl^{-}/Cl_2 reference electrode was calculated at the temperature range 573-723 K in fused LiCl-KCl-CsCl eutectic by *emf* method:

$$E^{*}_{Nd^{3+}/Nd^{2+}} = -(3.855 \pm 0.016) + (8.7 \pm 0.2) \cdot 10^{-4} \cdot T \pm 0.003, \quad V \qquad (4)$$

Basic thermodynamic properties of the reaction (5) were calculated and presented in Table:

$$NdCl_{2(l)} + ½\,Cl_{2(g)} \Leftrightarrow NdCl_{3(l)}. \qquad (5)$$

Basic thermodynamic properties of neodymium compounds in fused LiCl-KCl-CsCl eutectic at the temperature range 573-723 K. Apparent standard redox potentials Nd^{3+}/Nd^{2+} are given in the molar fraction scale.

Table

Thermodynamic properties	**573 K**	**623 K**	**673 K**	**723 K**
E^*/V	–3.356	–3.313	–3.269	–3.226
$\Delta G^*/(\mathrm{kJ\cdot mol^{-1}})$	–323.9	–319.7	–315.5	–311.3
$\Delta H^*/(\mathrm{kJ\cdot mol^{-1}})$	–372.0			
$\Delta S^*/(\mathrm{J\cdot K^{-1}\cdot mol^{-1}})$	84.0			
$K^*_{eq.}$	$3.46\cdot10^{29}$	$6.54\cdot10^{26}$	$3.14\cdot10^{24}$	$3.16\cdot10^{22}$
P_{Cl_2}/P_0	$8.35\cdot10^{-60}$	$2.33\cdot10^{-54}$	$1.01\cdot10^{-49}$	$9.98\cdot10^{-46}$

ELECTROCHEMICAL AND THERMODYNAMIC PROPERTIES OF THULIUM IN MOLTEN CHLORIDES

A. Novoselova, V. Smolenski
Institute of High-Temperature Electrochemistry UD RAS, Russia

Abstract

Actinides (An) recycling by separation and transmutation are considered worldwide as one of the most promising strategies for more efficient use of the nuclear fuel as well as for nuclear waste minimization, thus contributing to make nuclear energy sustainable. Pyrochemical separation techniques offer some potential advantages compared to the hydrometallurgical processes to separate An from lanthanides (Ln) contained in the irradiated fuel.

The goal of these investigations is to determine the electrochemical and thermodynamic properties of thulium compounds in molten alkali metal chlorides at large temperature region by transient electrochemical technique and *emf* method.

It was established that on an inert electrode Tm^{3+} ions are reduced to metallic thulium through two consecutive steps:

$$Tm^{3+} + \bar{e} \rightarrow Tm^{2+} \quad (1)$$

$$Tm^{2+} + 2\bar{e} \rightarrow Tm \quad (2)$$

The electroreduction of Tm^{3+} to Tm^{2+} ions was found to be reversible at low scan rates being controlled by the rate of the mass transfer and irreversible at high scan range ($> 0.1\ V\ s^{-1}$) being controlled by the rate of charge transfer.

The diffusion coefficient of *Tm(III)* complex ions was determined at different temperatures. Arrhenius law was verified by plotting the variation of the logarithm of the diffusion coefficient *vs.* reverse temperature.

The results of a study of the Tm^{3+}/Tm^{2+} couple redox potentials *vs*. Cl^{-}/Cl_2 reference electrode at wide temperature range in molten chlorides was carried out by direct potentiometric and cyclic voltammetry methods. Apparent standard redox potentials of the couples $E^{*}_{Tm^{3+}/Tm^{2+}}$ were determined in different solvents. Basic thermodynamic properties of the reaction (3) were calculated

$$TmCl_{2(l)} + \tfrac{1}{2} Cl_{2(g)} \Leftrightarrow TmCl_{3(l)}. \quad (3)$$

The comparison of the base thermodynamic properties of Tm compounds in molten alkali metal chlorides at 973 K. Apparent standard redox potentials Tm^{3+}/Tm^{2+} are given in the molar fraction scale.

Thermodynamic properties	**NaCl-KCl**	**NaCl-KCl-CsCl**	**NaCl-2CsCl**	**CsCl**
E^*/V	– 2.529	– 2.721	– 2.733	– 2.822
$\Delta G^*/(kJ\cdot mol^{-1})$	– 244.1	– 262.6	– 263.6	– 272.3
$\Delta H^*/(kJ\cdot mol^{-1})$	– 340.0	– 354.1	– 364.2	– 388.8
$\Delta S^*/(J\cdot K^{-1}\cdot mol^{-1})$	98.5	94.5	103.2	119.7
$K^*_{eq.}$	$7.76\cdot10^{14}$	$1.31\cdot10^{14}$	$1.44\cdot10^{14}$	$4.40\cdot10^{14}$
P_{Cl_2}/P_0	$6.11\cdot10^{-27}$	$5.86\cdot10^{-29}$	$3.50\cdot10^{-29}$	$5.61\cdot10^{-30}$

STRUCTUREAL STUDIES ON EXTRACTED SPEICES BY USING DIAMIDE COMPOUNDS AS EXTRACTANT FOR ACTINIDE/LANTHANIDE SEPARATION

Shin Okumura[1], Zoltán Szabó[2], Yasuhisa Ikeda[1]
[1]Tokyo Institute of Technology, Japan [2] KTH -Royal Institute of Technology, Sweden

Abstract

Diamide compounds have been examined as extractants for separating actinide(III) and lanthanide(III) from high level liquid waste (HLLW) by solvent extraction. *N,N,N′,N′*-tetraoctyl-glycoldiamide (TODGA) and *N,N,N′,N′*-tetraoctyl-3,6-dioxaoctanediamide (DOODA) have been reported to extract trivalent metal ions with large *D* values into *n*-dodecane from nitric acid solutions. TODGA is one of the most extensively studied and promising extractant. Recently, it was reported that DOODA also has similar *D* values and shows different affinity to heavier lanthanide(III). In the most of extraction of metal ions from aqueous phase to organic phase, metal ions are extracted through the formation of uncharged complexes with the extractant and anions such as nitrate. The number of extractants and nitrate ions participating in solvent extraction, which are usually obtained from *logD-log[L]* plots ([L] : the concentrations of the extractant or nitric acid), are not always compatible with those in the crystal structures. Several structural studies were made in this area, however the nature of extracted complexes is still not elucidated. In order to understand the extraction mechanism and the factors for controlling selectivity, the stoichiometry and the structure analysis of the extracted species in the solution should be essential.

In this paper, in order to elucidate the structures of the extracted species, the complex formation with lanthanide(III)-diamide were studied. Multinuclear NMR spectroscopy was used to obtain information about the stoichiometry, the symmetry and the structural parameters of lanthanide(III)-diamide complexes in acetonitrile. ^{1}H-, ^{13}C-, and ^{15}N-NMR signals were assigned by using 2D COSY and HMQC correlation spectroscopies and nuclear Overhauser effects (NOE). By increasing the metal to ligand ratios from 2:1 to 1:10. In the DOODA system, when the ratio metal/DOODA is less than 1, the ^{1}H NMR spectra showed that the metal and DOODA form 1:1 complex, and DOODA retains C2v symmetry in the 1:1 complex in the solution. Broad signals appeared between free DOODA and coordinated in the 1:1 complex when the ratio is more than 2. This indicate that the existence of a fast ligand exchange process on the NMR timescale. Paramagnetic metal ion complexes gave separate broad signals of free and coordinated DOODA, and the signals were resolved at lower temperature. The signals assigned to two coordinated DOODA molecules suggest that two DOODA are in symmetric positions but both of them coordinate asymmetrically. This result is in agreement with the solvent extraction result which has shown two DOODA molecules participate in the extracted species. The results of the TODGA system are also discussed from the viewpoint of the stoichiometry and the symmetry of the extracted species. Our study demonstrates that the structure analyses of the extracted species in the solution by using NMR can interpret well solvent extraction mechanism of lanthanide(III) by diamide compounds as an extractant.

Thermodynamic properties of heterogeneous alloys of lanthanum with gallium-indium eutectic

Schetinskii A.V.[1]**, Volkovich V.A.**[1]**, Yamschikov L.F.**[1]**, Dedyukhin A.S.**[1]**, Maisheva A.I.**[1]**, Osipenko A.G.**[2]**, Raspopin S.P.**[1]
[1] Ural Federal University, Ekaterinburg, 620002, Russian Federation
[2] JSC “State Scientific Center Research Institute of Atomic Reactors”, Dimitrovgrad, 433510, Russian Federation

Abstract

Currently, much attention is paid to the development of nuclear energy, including the fast breeder reactors. Using of these reactors will significantly increase the fuel burnup and reproduction of fissile materials. Pyrochemical methods are developed for processing spent nuclear fuel (SNF) of such reactors. Some of them are based on the use of molten salt mixtures. The main advantage of these methods is the high radiation resistance of melts. For separation of fuel components can be used deposition processes at the liquid metal of fuel elements or fission products. To carry out these processes is necessary to know the behavior of all components of these systems, including lanthanum, which is one of the fission products.

It is known that the gallium is one of the most useful metals for separation of uranium and lanthanum. The main reasons for this behavior of gallium - it's high rates of partition coefficients of U and La (thermodynamic estimate shows that in the temperature range 800-1000 K it varies from $1,4\cdot10^{4}$ to $5\cdot10^{5}$, which is significantly higher than for other low-melting metals) . To lower the melting temperature of the metallic phase should use its alloys. The lowest melting temperature in systems based on gallium-served observed for the alloy Ga - In (Tm = 289 K). In addition, we must remember that the eutectic metal systems in their behavior tend to of an ideal, so the vapor pressure over melts of Ga-In can be estimated from the additivity rule and calculated at 1100 K, the equilibrium vapor pressure above the eutectic melt of Ga - In is $1,14\cdot10^{7}$ atm. The use of alloys of this composition would, inter alia, to reduce the operating temperature of components separating process of the spent nuclear fuel.

Thus, a wide range of liquid states and low vapor pressure in the Ga - In system favor the development of processing of spent nuclear fuel at low temperatures using data from bimetallic melts. Working with the spent fuel in such an environment will allow a substantial but to simplify the of deep fractionation of nuclear waste in the system "liquid chloride salt - the metal" to achieve a return of 99.9% in the cycle of nuclear materials and training programs to the final disposal of waste.

The selectivity of the separation process is influenced by both properties of metal and salt phases at the interface which, passing through the separation of the chemical elements. Therefore, to study the thermodynamic features of eutectic alloy Ga - In in processing of irradiated nuclear fuel is necessary to study the thermodynamic properties of all components of the fuel, as in metal-ray, and in salt melts. There are literature data of the lanthanum behavior in binary metallic systems with gallium and indium. Records of the thermodynamic properties of lanthanum in the three-component metallic systems, including those consisting of Ga and In are not found, In the present study was an

experimental determination of the lanthanum activity in the eutectic alloy Ga-In in the temperature range 573 1073 K using the EMF.

As a result of this work were first identified lanthanum activity in the eutectic alloy Ga-In in the range 573-1073 K. It is shown that the saturated two-phase systems of lanthanum in equilibrium with the liquid phase intermetallic compounds are $LaGa_6$ (750 K) and $LaGa_2$ (after 750 K). Clarified the thermodynamic characteristics of two-phase alloys of lanthanum with gallium and indium, and lanthanum with indium-floor chennye earlier in a narrow temperature range.

BENCHMARK ON CU-NI CO-DEPOSITION IN $CUSO_4$/$NISO_4$ SOLUTION BY THREE-DIMENSIONAL MASS TRANSPORT AND ELECTROCHEMICAL MODEL

Jaeyeong Park[1*], Sungjun Son[1], Sungyeol Choi[1], Kwang-Rag Kim[2], Il Soon Hwang[1]
[1]Department of Nuclear Engineering, Seoul National University, 1 Gwanak-ro,
Gwanak-gu, Seoul 151-742, Republic of Korea
[2]Korea Atomic Energy Research Institute, 1045 Daedeok-daero, Yuseong-gu,
Daejeon, 305-353, Republic of Korea

Abstract

Highly radioactive isotopes remain within spent nuclear fuels even one million years after discharge from the reactors. To end this problem in a manageable period, pyroprocessing has explored to achieve sufficient throughput with inaccessible high temperature and strong radiation. Such remote operations, especially at uranium electrorefining, are challenges for repeated experiments that also generate a pile of radioactive wastes. Computational analysis is a plausible option to reduce trials and errors to find optimized designs and operating conditions of the electrorefiner.

To allow simulations to replace electrorefining experiments to the extent possible, we have developed a three-dimensional multi-species electrochemical model. Commercial computational fluid dynamics software was coupled with user-defined electrochemical modules. Ion transport equation governs overall mass transport and concentration distribution while concentration-modified Butler–Volmer equation controls electrochemical reactions at electrodes. This model was already validated by molten-salt electrorefining experiments using EBR-II spent fuels in cooperation with the Idaho National Laboratory. At that time, we benchmarked 80 hours long variations of overall cell potential only due to a lack of position-dependent information. This, it is needed to examine the model’s capability to predict position-dependent potential and current.

Here, we further validated the model with well-proven aqueous systems which experimentally measured more specific results other than cell poentential. Cu-Ni co-depositions in $CuSO_4$ and $NiSO_4$ solution were experimented by using both a corrosion cell and a rotating cylindrical Hull cell. First, transfer coefficients, exchange current densities, and other materials properties of Ni^{2+} and Cu^{2+} in sulphate solution was estimated by cathodic potentiodynamic polarization at the static corrosion cell. Second, the co-deposition experiment was galvanostatically conducted without stirring in the corrosion cell. Third, the co-deposition was also experimented in the rotating Hull cell. Fourth, simulated cathodic overpotential changes were validated to measured results at the corrosion cell. Fifth, cathodic overpotential distributions at the Hull cell were benchmarked with computational results, as a function of position, cathode rotating speeds, and applied current densities.

Benchmark tests at various operating conditions and two different geometries generally showed good agreement between simulations and experiments. Despite minor discrepancies at detailed potential variations, these could be improved by using more accurate electrochemical properties and considering current efficiency, nucleation overpotential, and other missing effects. Therefore, the multi-species co-deposition model could be a computational design tool to predict current densities and overpotentials for electrorefiners.

Purification of Used Zr Scrap by Molten Salt Electrorefining

Kyoung-Tae Park[1], Tae-Hyuk Lee[2], Wan-Kyu Kim[2], Eun-Kyu Lee[2], Soon-Ig Hong[2], Nam-Chan Jo[3], Jong-Hyeon Lee[1, 2]
[1]Graduate School of Green Energy Technology, Chungnam national university, Korea
[2]Department of Nano Material Engineering, Chungnam national university, Korea
[3]KEPCO NF Co., Ltd., Korea

Abstract

Zr and its alloys are one of the most important materials used as cladding nuclear fuels because of its high dimensional stability, good corrosion resistance and especially, lowest neutron-absorbing cross section. However, manufacturing costs of nuclear grade Zr from Zircon ore is very high because satisfying compositional regulation is very difficult, which included separation of impurities, using multiple solvent extraction and distillation. And also, purification of used Zr demands same processes compared with zircon ore refining process. In this study, Zr scrap was recycled by molten salt electrorefining instead of commercial process. For pure Zr production from Zr scrap, electrochemical cut-off potential was controlled by applied current density, during the this process noble metal impurities such as Nb, Sn Fe and etc. drop down to the bottom of electrolyte cell. Corrected Zr deposits were analysed by SEM, XRD, TGA, ICP-MS. As a result of molten salt electrorefining, composition of Zr was limited under ASTM B349, nuclear grade Zr sponge specification.

REDOX REACTIONS OF NEPTUNIUM WITH NITROUS ACID AND ACETOHYDROXAMIC ACID UNDER RADIOLYSIS CONDITIONS

Martin Precek[1], Alena Paulenova[1], Bruce Mincher[2]
[1]Oregon State University, Corvallis, USA
[2]Idaho National Laboratory, Idaho Falls, USA

Abstract

Nitrous acid (HNO_2) is an important redox-active compound significantly influencing the extraction behavior of neptunium and plutonium during separation processes. Nitrous acid, present in high content in the aqueous phase, acts as a stabilizer of extractable tetravalent plutonium; it has a catalytic effect on the rate of oxidation of Np(V) by HNO_3, but on the other hand, it also leads to the reduction of a significant portion of neptunium to its non-extractable pentavalent state. Redox behavior of neptunium in gamma-irradiated nitric acid of 0.5 M – 4 M concentration and radiolytic production of HNO_2 was investigated spectrophotometrically.

Nitrous acid was found to be well extracted (log D≈ 1). The equilibrium between Np(V) and Np(VI) was achieved regardless of the initial fractions of the oxidation states Np(V):Np(VI), which varied from 5:95 to 45:55, and the final redox speciation of neptunium was found to be controlled primarily by the radiolytically produced nitrous acid. The radiolytic yields of HNO_2 in the aqueous phase were found significantly lower than in the organic phase – which is a result of much smaller presence in the organic phase of nitrite-scavenging intermediate species produced by the radiolysis of water (such as the hydroxyl radical or hydrogen peroxide).

Nitrous acid is also scavenged by acetohydroxamic acid (AHA, $CH_3CO\text{-}NHOH$) that was proposed as a salt-free stripping reagent to advanced PUREX process (UREX) to effectively separate neptunium and plutonium from uranium product. While rapidly reducing Np(VI) and Pu(VI), and forming un-extractable complexes with Np(IV) and Pu(IV), AHA doesn't affect the extraction of uranium. However, investigation of the redox stability of Np(V) in solutions of AHA-HNO_3 has revealed that after several hours, once all AHA is hydrolyzed into hydroxylamine, nitrous acid generated by autocatalytic reaction of hydroxylamine with nitric acid causes a sudden reoxidation of Np(V) to Np(VI). This effect prevents utilization of AHA as a long-term holding reductant for neptunium and plutonium and can complicate reprocessing operations.

OPTIMIZATION OF THE EXTRACTION OF ^{137}Cs AND ^{90}Sr FROM HIGH LEVEL LIQUID WASTE

Jiří Rais,[a] Shoichi Tachimori,[b] Jana Alexová,[a] Mária Bubeníková[a]
[a] Nuclear Research Institute, plc, Řež, 25068 Husinec – Řež 130, Czech Republic
,
[b] Secretariat of Nuclear Safety Commission of Japan, Tokyo, 100-8970 Japan

Abstract

It is anticipated that in future, when nuclear waste repositories will be constructed in many countries, the separation of individual components and their separate handling or burial will become urgent. This persuasion stems from options of retrievability, individual burying, and possible further use of individual radionuclides.

The two main processes for the extraction separation of the title radionuclides are so called “dicarbollide process” (developed in NRI and industrially used in Russia) and “FPEX process” (being developed in USA). The two reference processes differ in used extraction agents, characteristics of extraction, and possibly in level of chemical and radiation stability. Whereas “dicarbollide process”, using highly stable dicarbollide anion, is to be viewed as a classical, new processes use cationic organic selective ligands, i.e. special crown ethers for extraction of ^{90}Sr and calixcrowns for ^{137}Cs extractions. The “dicarbollide process” was in detail studied before in NRI and our institute participated also actively in development of selective ligands for extraction of Cs in the frame of Japanese ARTIST project proposed and developed by one of us (S.T.)

The main obstacles in both types of processes lie in the choice of suitable solvent. In this report, we present some results for two ecological variants of dicarbollide solvents. Further, we compare and evaluate some alcohol solvents for systems with selective ligands and extraction of ^{137}Cs and ^{90}Sr.

The studies were financially supported by Grant Agency of Czech Republic, grant No. P106/11/2319

Minor actinide partitioning: main outcomes in the framework of the 2006 French act

**Christine Rostaing, Christophe Poinssot, Dominique Warin, Pascal Baron,
Nathalie Herlet**
Commissariat à l’énergie atomique et aux énergies alternatives, Marcoule,
France

Abstract

The Waste Management Act passed by the French Parliament on June 28, 2006, demands that Partitioning and Transmutation research are conducted in strong connection to GEN IV systems and ADS development, allowing to assess the industrial perspectives of such systems in 2012.

In this framework, the CEA had launched an important program which aimed at :

- developing different hydometallurgical processes for minor actinide separation depending on the scenario selected for the future nuclear cycle : homogeneous recycling (Pu, Np, Am, Cm) heterogeeneous (Am,Cm or Am alone) recycling,
- getting closer to process implementation conditions to assess industrial feasability of the various concepts developped.

Between 2007 and 2010, three concepts have been developped and validated in the Atalante facility through counter current hot tests implemented in mixer-settlers, using some hundred grams of spent fuel :

- GANEX for Grouped ActiNide Extraction is dedicated to homogenoeus recycling. It consists of two steps : the first step uses a monoamide extractant for the selective extraction of uranium. The second step is an adaptation of the DIAMEX-SANEX process. The solvent is a mixture of DMDOHEMA and HDEHP diluted in HTP. The transuranium elements (Pu, Np, Am, Cm) are coextracted with lanthanides and then selectively stripped with a polyaminocarboxylic acid (HEDTA for example) in a buffer medium at pH around 3.
- SANEX-TODGA is fitted for the heterogeneous recycling of Am+Cm in blankets put in the periphery of the reactor. The solvent is a mixture of TODGA (a diglycolamide) and TBP in http. After coextraction of An(III) and Ln(III), An(III) are selectiveley stripped with DTPA in a buffered malonic acid solution at pH 2.5.
- EXAm is adapted for Am alone recovery. The key is the extraction-scrubbing step : Am and light lanthanides are extracted while Cm and heavy lanthanides are recovered in the raffinate thanks to the diamide in the organic phase combining with a complexing agent (TEDGA) in the aqueous phase.

In the same time, studies have been conducted to assess industrial feasibility. The processes based on liquid-liquid extraction benefit from the experience gained by operating the PUREX process at the La Hague plant. Nevertheless, the processes present specificities which have been considered: evolution of the simulation codes, piloting of the processes, definition of the implementation conditions in continuous contactors; stability and solvent clean-up studies to

assess long term behavior of the separation systems towards radiolysis and hydrolysis, liquid waste management.

Significant results issuing from this research work are given.

Electrochemistry of selected lanthanides in LiF-BeF_2 system

Martin Straka, Lórant Szatmáry, František Lisý
Department of Fluorine Chemistry, Nuclear Research Institute Řež, Czech Republic

Abstract

Electrochemical based separation of actinides and lanthanides from molten salt media seems to be suitable method for separation of fissile material and fission products within the proposed fuel cycle of Molten Salt Reactor (MSR). To manage the reprocessing part of MSR fuel cycle, it is necessary to have the knowledge electrochemical behaviour of supposed fissile material and fission products in molten salts media. This work focuses on electrochemical of selected lanthanides in LiF-BeF_2 system which is considered to be a primary choice for MSR system. Characterization of LiF-BeF_2-Ln systems was done by cyclic voltammetry and chronopotentiometry. Comparison with similar BeF_2-free systems was done in order to describe the influence of specific characteristics of BeF_2 (BeF_2 is highly viscous, network-forming component). Diffusion coefficients of lanthanide ions were calculated where possible. Electrolytic experiments were done to evaluate the possibility of the deposition on several electrode materials (W, Mo, Ni). Alloying processes were recognized and the deposits were analyzed by XRD and SEM analysis. Again, the influence of Be present in the melt was recognized.

SEPARATION OF MINOR ACTINIDES AND FISSION PRODUCTS BY USING SOLID EXTRACTANTS

Tatsuya Suzuki[1], Yu Tachibana[1], Shin-ichi Koyama[2] Masanobu Nogami[3]
[1]Nagaoka University of Technology, [2]Japan Atomic Energy Agency, [3]Kinki University, Japan

Abstract

The solid type extractants are able to be applied to the chromatographic technique. The chromatographic technique is appropriate for the separation methods which there are many kinds of requiring elements i.e., the nuclide separation from spent nuclear fuels. We have been studied the separation of trivalent minor actinides and many kinds of fission products. The pyridine resin, crown ether resin, chelating resin, a kind of ion exchanger, and so on is used as solid type extractans. The pyridine resin is used for the separation of minor actinides from lanthanide, the mutual separation of minor actinides, the recovery of platinum group elements, etc. the crown ether resin is used for the separation of alkaline earth elements. Ferrocyanides, the adsorbent with carbonate ion and others have been also tested. In this report, the separation methods in the situation of many cases are described. One is the nuclide separation in the newly developed reprocessing process “Adv. Orient cycle”, which process is base on hydrochloric solution. One is the nuclide separation adding the PUREX or advanced aqueous reprocessing processes. One is the minor actinide separation from the molten salt waste. Another is the separation of radio elements from aqueous waste with sea water.

This work was supported by Grant-in-Aid for Scientific Research (B), MEXT (No.23360423)

PULSE ELECTROLYSIS OF URANIUM IN MOLTEN FLUORIDE SALTS

Lórant Szatmáry, Martin Straka, František Lisý
Department of Fluorine Chemistry, Nuclear Research Institute Řež, Czech Republic

Abstract

Knowledge of basic electrochemistry of uranium and study of its electrodeposition in molten salts is important for pyrochemical concepts of spent nuclear fuel reprocessing including “online” reprocessing concept for Molten Salt Reactor (MSR). The electrodepositions of uranium in molten fluoride salts (LiF-NaF-KF, LiF-CaF_2) were carried out under current pulses in the melt. Nickel working electrodes were used and alloying effects were observed. Compared to potentiostatic control of electrolysis, the deposit seems to be more compact, with significantly lower amount of carrier melt in it. The deposits were characterized by XRD and SEM-EDX methods. However, proper analysis with differentiation of several forms of the uranium was not possible and therefore it was not possible to make quantitative conclusions.

ALTERNATIVE PYROCHEMICAL REPROCESSING OF USED NUCLEAR FUEL USING MOLYBDATE MELTS

Peter Tkac, Terry A. Cruse, James L. Willit, and Mark A. Williamson
Argonne National Laboratory, USA

Abstract

Reprocessing of used nuclear fuel using molybdate melts containing molybdenum trioxide (MoO_3) and sodium molybdate (Na_2MoO_4) is an interesting alternative to chloride/fluoride-based pyrochemical processes. All U, TRU and the majority of the fission products are soluble in the molybdate melt. Through proper control of the salt composition, uranium can be selectively precipitated while fission products remain in the melt. The separation process based on the mixture of MoO_3 and Na_2MoO_4 is achieved because the fission products are much more soluble in the molybdate melt than UO_2. Moreover, the very low solubility of UO_2 in Na_2MoO_4 can be used for additional purification from most of the fission products. Small-scale experiments using gram quantities of uranium have been performed. X-Ray diffraction was used to identify the major uranium species that are formed at various salt compositions. Initial data for separation factors with respect to selected fission products in uranium product will be reported.

Kinetics back-extraction of Tc (VII) by DFG from 30% TBP solution using Lewis cell

Vitaly Vidanov, Konstantin Dvoyeglazov, Vladimir Volk
JSC “Academician A.A. Bochvar High-Tech Research Institute of Inorganic Materials”,
5a, Rogova str., Moscow, Russia

Abstract

In the PUREX process, which primarily used for uranium and plutonium separation, significant attention is paid to deep uranium purification from transplutonium elements and fission products, particularly from the technetium.

The uranium purification from technetium is one of the most intricate tasks in the process of spent nuclear fuel (SNF) reprocessing.

The uranium recycling, which is implemented through the separation process, requires the uranium reclaim be highly decontaminated from technetium in order to avoid problems arising during the volatile compounds of Tc formation at the sublimation stage.

In this study the kinetics back-extraction of Tc (IIV) from 30% TBP/dodecane into nitric acid solution containing diformil hydrazide (DFG) using Lewis cell was performed. The different parameters the back-extraction rate of Tc(VII) such as nitric acid, DFG, Tc concentration, influence of temperature and stirring speed were separately studied.

ALPHA-RAY IRRADIATION ON ADSORBENTS OF EXTRACTION CHROMATOGRAPHY FOR MINOR ACTINIDE RECOVERY

Sou Watanabe, Sachiko Miura, Yuichi Sano, Kazunori Nomura, Yoshikazu Koma and Yasuo Nakajima
Japan Atomic Energy Agency, Japan

Abstract

Extraction chromatography technology is one of the promising methods to recover minor actinides (MA; Am and Cm) from spent FBR fuel, and Japan Atomic Energy Agency has been conducting R&D study for the implementation. In order to design operational condition and frequency of adsorbents exchange, durability of the adsorbents is indispensable information. Although effects of γ-ray irradiation and exposure to acid on adsorption/elution performances of the adsorbents have been investigated so far, resistance against α-ray irradiation has not been reported yet. In this study, α-ray irradiation experiments on the representative adsorbents for the extraction chromatography process (CMPO/SiO_2-P, TODGA/SiO_2-P, HDEHP/SiO_2-P and isoHex-BTP/SiO_2-P) were carried out and influences of alpha-ray irradiation on fundamental characteristics of the adsorbents were investigated.

Nitric acid solutions containing ^{241}Am with 1.0 ~ 10 mM was prepared as α-ray sources. 0.5 g of the adsorbent was contacted with 10 mL of the source solution and was shaken for 3 hours to adsorb ^{241}Am onto the adsorbents. The ^{241}Am bearing particles were left for a certain period to irradiate with the α-ray. After the irradiation, ^{241}Am was desorbed by contacting the adsorbent with eluents. Batch adsorption/elution experiments using the irradiated adsorbents were carried out to evaluate influences of α-ray irradiation on distribution coefficients and desorption ratio. DG/DTA analysis on the α-ray irradiated adsorbents was also carried out to examine a possibility of hazardous reaction due to the formation of radiolysis products.

For all adsorbents, distribution coefficients and desorption ratios of ^{241}Am decreased with increasing the radiation dose, where the radiation dose was calculated from the amount of ^{241}Am adsorbed with an assumption of that α-ray energy from ^{241}Am (E = 5.5 MeV, Half life = 432.2 y) is completely transferred to the adsorbent. Those decreases in the performances must be corresponding to degradations of extractants impregnated in the adsorbents and of polymers supporting the extractants. Comparing the influences of α-ray with γ-ray on those properties under the same radiation doses condition, radiation damages suffer from the α-ray irradiations are less significant for all the adsorbents. Difference between a charged particle and γ-ray is considered to cause the different interaction between the system (adsorbents with nitric acid) and the radiations. Detailed mechanisms of the radiolytic degradations will be discussed in the presentation.

Distinct exothermic peaks were not observed at under 100°C in the DTA curves. In the extraction chromatography system, temperature of the system is controlled at less than 100°C to prevent from boiling the mobile phase. Therefore, degradation which is suspected to lead abnormal events such as fire or explosion will not be anticipated for the use of the adsorbents.

Posters - Session VI

CURRENT STATUS AND FUTURE DEVELOPMENTS OF THE TRANSURANUS CODE: OXIDE FUELS FOR MINOR ACTINIDES RECYCLING

R. Calabrese*, V. Di Marcello^, A. Schubert^, J. van de Laar^, P. Van Uffelen^
*ENEA Agenzia nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile, Italy
^European Commission, Joint Research Centre, Institute for Transuranium Elements, Germany

Abstract

Nuclear energy thanks to its competitiveness and low carbon technology is playing a significant role in the global energy market. While nowadays nuclear plants are almost entirely based on thermal reactors technology, a new generation of fast reactors is investigated pursuing an improvement of the effectiveness in the use of natural uranium resources in parallel with an increase in proliferation resistance and a reduction in radiotoxicity of used fuel, achieved through the transmutation of minor actinides (MA). These innovative nuclear plants, conceived in the framework of the Generation IV International Forum (GIF), have a deployment time horizon beyond 2030.

The transmutation of minor actinides is achieved through two main options: homogeneous recycling, where small quantities of MA are diluted in the driver fuel (MOX), and heterogeneous recycling, where MA are dispersed in high concentration in a UO_2 matrix and irradiated at the periphery of the core. The addition of minor actinides in oxide fuels has an impact on various aspects of in-pile performance such as the degrading of thermal conductivity and melting temperature, a higher fuel swelling rate and a higher helium production. These aspects need to be carefully studied to fulfill safety criteria as a key requirement to support the expansion of nuclear business. The description and modelling of MA-bearing oxide fuels is therefore challenging and investigations are ongoing in various projects undertaken in the EURATOM framework e.g. FAIRFUELS, PELGRIMM.

With regard to fuel with a standard fabrication route, the paper discusses the current status and future modelling needs of the TRANSURANUS fuel performance code, with particular attention to following aspects:

- helium generation;
- integral fission gas release of xenon, krypton and helium;
- solid and gaseous fuel swelling;
- fuel thermal conductivity;
- re-distribution of plutonium at high temperatures.

IRRADIATION PERFORMANCE COMPARISON OF EXPERIMENTAL AND PROTOTYPIC LENGTH METALLIC (U-10ZR) FUEL

Jon Carmack, Douglas Porter, Heather Chichester, and Steven Hayes,
Idaho National Laboratory, P.O. Box 1625, Idaho Falls, Idaho 83415-6188,
jon.carmack@inl.gov

Abstract

Metallic fuelled fast reactors have demonstrated excellent operational and safety performance. Sodium cooled fast reactors are candidate systems for accomplishing the transmutation of minor actinide elements. Two fast reactors, the Experimental Breeder Reactor II (EBR-II) and the Fast Flux Test Facility (FFTF) have tested and demonstrated the performance of metallic (U-10Zr) alloy fuel. The EBR-II reactor featured a 13.5 inch core height and the FFTF a 91.4 cm core height. This paper explores the steady state performance differences between these two core sizes through the X447 EBR-II experiment and the MFF-3 and MFF-5 assembly irradiations in FFTF. The experiments conducted in the EBR-II and the FFTF provide invaluable data on the performance and behavior of metallic nuclear fuels. Understanding the behaviours that may be impacted by fuel column height is needed to accurately predict fuel pin and assembly performance and provide the reactor designers with the data needed when increasing fuel pin height.

The X447 experiment was conducted in the EBR-II as part of the Integral Fast Reactor program’s development and testing of metallic fuel. After achieving a burnup of 5 at.% (288 EFPD), the assembly was removed from the reactor, reconstituted as X447A, and reinserted into the reactor. No fuel pin failures were detected or observed at this interim burnup but four fuel pins were selected from the assembly and subjected to postirradiation examination. After re-insertion into EBR-II, the X447A experiment achieved 10 at. % BU (641 EFPD) and was removed from the reactor. The experiment was designed to operate at high temperature (630°C to 660°C PICT). The high cladding temperatures were expected to challenge the creep rupture resistance of the cladding alloy, HT9, as fission gas pressure increased and FCCI thinned the cladding wall. Two fuel pin breaches occurred at approximately 10 at.% burnup.

The MFF series of metallic fuel irradiations [1] provides an important potential comparison between fuel performance data generated in EBR-II and that expected in a larger scale fast reactor. The FFTF reactor contained standard fuel with a 91.4 cm (36-inch) tall fuel column and a chopped cosine neutron flux profile resulting in a core with peak cladding temperature at the top of the fuel column but with peak burnup near the centerline of the core. The peak fuel centerline temperature was midway between the core center and the top of fuel, lower in the fuel column than that in the EBR-II fuel pins. The MFF-3 and MFF-5 qualification assemblies operated in FFTF to greater than 10 at. % burnup with no pin breaches. The MFF-3 assembly operated to 13.8 at.% burnup with a peak inner cladding temperature of 643°C. The MFF-5 assembly operated to 10.1 at.% burnup with a peak inner cladding temperature of 651°C. This can be compared to the two pin breaches experienced in X447 at approximately 10 at.% burnup and peak inner cladding temperatures of 648°C and 638°C respectively. A selection of the X447 postirradiation examination was published in 1993 [2]. Comparison of the X447 postirradiation and the MFF post irradiation indicates that fuel behavior is similar between long and short fuel

column heights and predictable when considering the flux, power, and temperature profiles of the fuel.

References

[1] Pitner, A.L. and Baker, R.B. “Metallic Fuel Test Program in the FFTF.” *Journal of Nuclear Materials*, v. 204 (1993) pp. 124-130.

[2] Pahl, R.G., C.E. Lahm, and S.L. Hayes, “Performance of HT9 Clad Metallic Fuel at High Temperature.” *Journal of Nuclear Materials*, v. 204 (1993) pp. 141-147.

Investigation of pelletized and sphere-packed oxide fuels for Minor Actinide transmutation in Sodium Fast Reactors, within the FP-7 European project PELGRIMM

F. Delage[1], S. Béjaoui[1], S. Lemehov[2], J. MacGinley[3], W. Maschek[4]
On behalf of the PELGRIMM European project consortium
[1]CEA -France, [2]SCK●CEN - Belgium, [3]JRC/ITU – Germany, [4]KIT – Germany

Abstract

The FP-7 European project PELGRIMM, which stands for "PELlets versus GRanulates: Irradiation, Manufacturing, Modelling" is a 4 year and 7.2M€ project, addressing Minor-Actinide (MA) bearing oxide fuel developments for Generation IV – Sodium Fast Reactor Systems. Both options, MA homogeneous recycle in driver fuels with MA content at a few percent and heterogeneous recycle on UO_2 fuels bearing high MA contents located in the radial core blanket, are considered. Two fuel forms: pellet and spherepac, are under investigation.

A total of 12 partners from research laboratories, universities and industries, collaborate to share and leverage their skills, progress and achievements, covering a comprehensive set of investigations:

- perform the Post-Irradiation Examinations of MARIOS and SPHERE irradiated pins within the European project FAIRFUELS, providing the very first results respectively on the helium behaviour in $(Am,U)O_2$ fuels and a comparison between sphere-packed and pelletized $(U,Pu,Am)O_2$ fuel performances;
- take the next step in the $(Am,U)O_2$ fuel qualification rationale by performing the semi-analytical test MARINE in HFR;
- extend Minor Actinide bearing fuel fabrication processes to alternative routes in order to limit secondary waste streams;
- extend the capabilities of existing calculation codes devoted to fuel behaviour under irradiation
- accomplish a preliminary design of a sodium cooled fast reactor core with spherepacked $(U,Pu,Am)O_2$ fuels and perform a preliminary safety assessment.

PELGRIMM promotes the implication of European students and young researchers too, through:

- placements of trainees in organisations involved in the project
- the development and delivery of training courses and workshops
- contributions to research actions of the project by researchers preparing a doctoral thesis or employed at a post-doctoral position.

The paper gives an overview of the PELGRIMM programme to be completed by December 2015.

RADIAL PROFILE OF THERMAL DIFFUSIVITY AT HIGH SPATIAL RESOLUTION ON METALLIC TRANSMUTATION FUELS

J. Rory Kennedy, Matthew K. Fig, Heather MacLean-Chichester
Idaho National Laboratory, USA

Abstract

In the realm of radioactive materials, a major challenge to the development of materials is the measurement of the properties for which the material is being developed. For example, the phenomenon of microstructure evolution of a nuclear fuel in reactor is well known but the details of the effects of the change on the behavior of such important phenomena as thermal conductivity, mechanical properties, and phase formation have not been quantified at high spatial resolution. There is a strong need to develop or adapt advanced instrumentation for measurements on radioactive materials. Idaho National Laboratory has an ongoing effort to develop or adapt a variety of measurement techniques to radioactive and highly radioactive materials. These efforts are also coupled with efforts to produce experimental results at spatial and temporal scales that are equivalent to those available to computational modeling and simulation. A laser based device called the Scanning Thermal Diffusivity Microscope, conceived and developed over the past few years, has recently been installed in a hot cell where examinations of fresh and irradiated fuel samples have begun in order to profile the thermal diffusivity of fuels and materials down to 50μm spatial resolution. Recent high spatial resolution thermal diffusivity results from the radial profiling of metallic transmutation fuels at various levels of burnup will be presented.

Posters -
Session VII

SENSITIVITY/UNCERTAINTY ANALYSIS OF THE VALIDATION OF THE EVOLCODE 2.0 BURN-UP SYSTEM WITH PWR EXPERIMENTAL DATA

F. Álvarez-Velarde and E. M. González-Romero
Nuclear Innovation Program
Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas
CIEMAT. Madrid (Spain)

Abstract

The prediction of the isotopic composition of irradiated nuclear fuels is crucial for studies on burn-up credit, reactor physics and waste management. With this motivation, CIEMAT has been developing, upgrading and validating for the last decade the burn-up simulation system EVOLCODE 2.0.

One of these validations involved the ICE experiment, a whole irradiation of some UO2 fuel assemblies in a pressurized light water reactor (PWR) for a total of around 30 GWd/tU and the later isotopic content experimental measure of U and Pu isotopes. The validation of EVOLCODE 2.0, consisting in the simulation of the PWR with a single pin model, provided very satisfactory results (with mass deviations between the simulation and the experimental data smaller than 3% for the main U and Pu isotopes), with generally smaller mass deviations than those obtained with the deterministic code KAPROS. However, for some actinides the obtained mass deviations are larger than the experimental uncertainties.

The aim of this work is to analyze the simulation uncertainties in the final isotope masses and to comprehend their possible sources. For this reason, we have performed an uncertainty analysis based on the sensitivity methodology, providing the uncertainties in the isotopic content propagated from the cross sections uncertainties, and the energy ranges and isotopes responsible of the major contributions to these uncertainties as well. The COMMARA data library has provided us with both the covariance information and the cross section uncertainties in 33 energy groups.

Preliminary results show that the cross section uncertainties are an important contributor to the simulation uncertainties, sometimes even larger than the experimental and the simulation uncertainties. Besides, some simulation uncertainties for actinides with very small amount are significantly larger than the other uncertainties and need further investigation.

This work is framed within the ANDES project (Accurate Nuclear Data for nuclear Energy Sustainability, 7th Framework Programme of the European Union). Its results will allow us to give some guidelines in order to advise additional strategies for cross section uncertainty reduction.

Thermal Hydraulic Study of Natural convection in the Heavy Eutectic liquid metal Loop HELIOS, Benchmark RESULTS on Lead-Alloy Cooled Advanced Nuclear Energy System (LACANES)

Abdalla Batta, Andreas Class
Karlsruhe Institute of Technology, Germany
Jae Hyun Cho, Il Soon Hwang
Seoul National University, (SNU) S. Korea
Paride Meloni, Saverio Nitti, Massimiliano Polidori
Italian National Agency for New Technologies, Energy and Sustainable Economic Development, Italy
Vincenzo Casamassima
Ricerca sul Sistema Energetico / Power Generation Systems Department, Italy
Yong Joon Choi
OECD Nuclear Energy Agency, Nuclear Science Section (OECD/NEA), France

Abstract

Liquid lead-bismuth eutectic (LBE) has been extensively investigated in worldwide because of their low melting temperature, high boiling temperature, chemical stability and neutron transparency. The study of natural convection heat transfer of liquid metals becomes very important for the safety of next generation nuclear reactors. Design tools validation like system codes and computational fluid dynamic (CFD) models requires reliable experimental data. Under the guidance of the Nuclear Science Committee (NSC) and the mandate of the Working Party on Scientific Issues of the Fuel Cycle (WPFC), the Task Force on the Benchmarking of Thermal-Hydraulic Loop Models for Lead-Alloy Cooled Advanced Nuclear Energy Systems (LACANES) is started. One of the objectives of this WPFC is the validation of thermal-hydraulic loop models for application to LACANES design analysis in participating organisations, by benchmarking with a set of well-characterised lead-alloy coolant loop test data. Other objectives are establishment of the guidelines for quantifying thermal-hydraulic modelling parameters related to friction and heat transfer by lead-alloy coolant and identification of specific issues, either in modelling and/or in loop testing. The Heavy Eutectic liquid metal Loop for Integral test of Operability and Safety of PEACER (HELIOS) which is constructed in Seoul National University of Republic of Korea in 2005 is considered for benchmarking purposes. The 12 m height loop is driven by natural convection induced by heating of the core at lower level of the loop and removing of the gained head at the heat exchanger placed in the upper part of the loop, see figure 1. In the natural convection case the pump is not activated. Bypass pipe is connected between core inlet and loop cold leg downstream of heat exchanger. The temperature values are measured at different location of the loop. Mass flow rate is also measured. Steady and unsteady operation conditions and data are generated.

In the current study results obtained by CFD and system codes are presented. Different heating modes in the core are simulated by CFD in order to demonstrate the importance of location of heat deposition in the loop. The various CFD results obtained for the core are compared to benchmark results obtained by different system codes. This validates the used heat transfer and pressure losses correlations of the system codes. Based on these data the thermal hydraulics investigation of the heat exchanger is assessed. The comparison of benchmark data to numerical data shows good capability of the used simulation tools for the studied natural convection case.

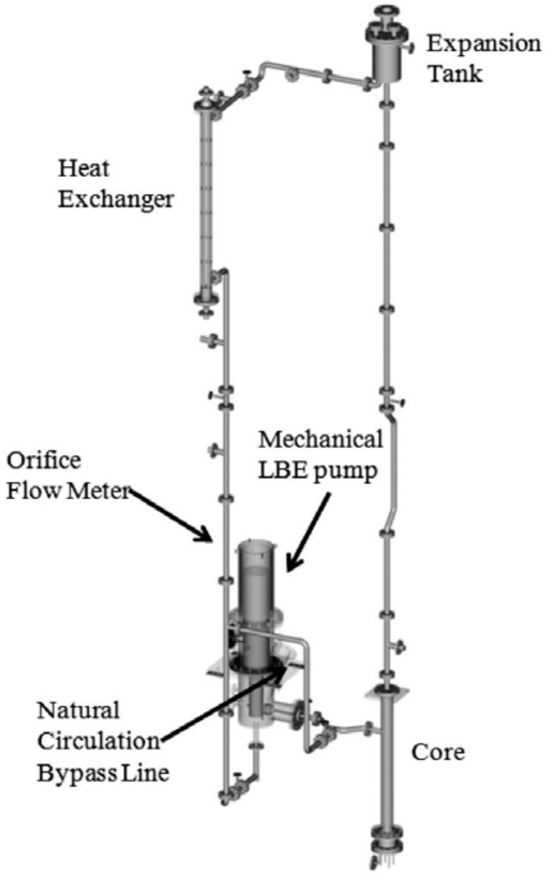


Figure 1. HELIOS Loop, Showing Main Loop Components

Accurate Nuclear data for present and future reactors from the ANDES project

E. M. González-Romero on behalf of the ANDES Euratom project
Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas
CIEMAT. Madrid (Spain)

Abstract

The ANDES FP7-EURATOM project, Accurate Nuclear Data for nuclear Energy sustainability, intends to address the nuclear data needs associated to the new reactors and new fuel cycles supported by SNETP, in its strategic research agenda and in the ESNII proposal, taking into account the priority lists for nuclear data from NEA/OECD, FP6-EURATOM projects EUROTRANS-NUDATRA and CANDIDE.

ANDES combines a reduced group of selected differential measurements, the improvement in uncertainties and covariance's within the evaluation process and the validation of present and new data libraries using integral experiments, to bring most critical nuclear data to the level of accuracies required by the new reactors and system promoted by ESNII and the SNETP. In addition, a specific work package will improve the prediction capabilities of high-energy transport codes for the design of ADS, developing better models and performing a few selected measurements.

The research of ANDES is particularly relevant for P&T research because the involvement of isotopes, materials and concepts not widely included in present reactors and often lacking of required accuracy and operational experience.

The progress and preliminary results of the project will be presented in this paper, including:

Accurate new measurements of capture and other cross sections for minor actinides like 241Am, major actinides like 238U, fission cross sections of Pu and minor actinides, and for other inelastic cross sections,

Recent developments on data evaluation tools, particularly to include uncertainty and covariance matrices, as well as updates of simulation tools to facilitate the regular use of these covariances in standard problems,

Comparisons of new calculation tools, data and codes, with criticality and experimental reactor benchmarks as well as with dedicated integral experiments, and feedback to evaluators and measurement programs,

Recent validation of high energy reaction models, new data from neutrons beyond 100 MeV and progress in the high energy reactions models for improving accuracy and extending the energy range of applicability.

Finally an outlook of the additional results expected by the end of the project will also be included.

Perspective of the Accelerator-Driven System with High-Energy Protons in the Kyoto University Critical Assembly

Cheol Ho PYEON, Takahiro YAGI and Tsuyoshi MISAWA, Hironobu UNESAKI and Ken NAKAJIMA
Nuclear Engineering Science Division, Research Reactor Institute, Kyoto University
Asashiro-nishi, Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan
E-mail: pyeon@rri.kyoto-u.ac.jp

Abstract

At the Kyoto University Research Reactor Institute, the experimental study on the accelerator-driven system (ADS) is being conducted with the use of the Kyoto University Critical Assembly (KUCA) and the fixed-field alternating gradient (FFAG) accelerator. The spallation neutrons generated by 100 MeV proton beams from the FFAG accelerator have been successfully injected into not only the uranium-loaded core but also the thorium-loaded core. A series of the thorium-loaded ADS experiments has been carried out by varying the external neutron source (14 MeV neutrons and 100 MeV protons) and the neutron spectrum of the core with the combined use of fuel (highly-enriched uranium: HEU, natural uranium: NU and thorium: Th) and moderator (polyethylene, graphite, beryllium and aluminum). In the thorium-loaded ADS experiments, the reactor physics parameters have been successfully obtained, including the reaction rate distribution, the neutron decay constants, the neutron multiplication and the subcriticality, with the use of the thorium capture reactions in subcritical systems.

The reactor physics parameters in the subcritical systems, including the neutron multiplication M ($M=(F+S)/S$, F: fission neutrons and S: source neutrons) and the subcritical multiplication factor k_s ($k_s=F/(F+S)$), could be evaluated successively by the experimental and numerical (MCNPX) reaction rates of the indium wire and foil obtained in the core and the target, respectively, as well as in previous study on the Th-loaded ADS, with the variation of the subcriticality, the external neutron source and the neutron spectrum. Further, the benchmark experiments on the Th-loaded ADS are planned to be carried out to conduct the conversion analysis of ^{232}Th capture and ^{233}U fission reactions with the use of capture and fission ratio: ^{232}Th/^{238}U (NU) and ^{233}U/^{235}U (HEU), respectively, in case of the variation of the subcriticality, the external neutron source and the neutron spectrum.

The U-loaded ADS experiments could be also conducted in the subcritical core with harder spectrum than previous cores to examine the feasibility of the nuclear transmutation with the use of ADS, including the minor actinides (MAs: ^{237}Np and ^{241}Am). Additionally, as well as shown in the Th-loaded ADS experiments, another conversion analysis of ^{237}Np/^{238}U and ^{241}Am/^{235}U is expected to be carried out to investigate the MAs characteristics when the subcriticality, the neutron spectrum and the external neutron source are varied in the subcritical system.

INTELLIGENT NUCLIDE SELECTION CAPABILITY IN THE REACTOR PHYSICS AND INVENTORY CALCULATION CODE SERPENT 2

Tuomas Viitanen and Jaakko Leppänen
VTT Technical Research Centre of Finland

Abstract

Serpent is a user-friendly reactor physics and inventory calculation code developed at VTT and used by many organizations around the world [1]. Since the neutron transport of Serpent is based on the continuous-energy Monte Carlo method, it is well-capable of calculating the nuclide inventories for all kinds of reactors and fuels – from ordinary LWRs to fast reactors – with high accuracy.

The output of Serpent 2 is versatile, containing for example total and nuclide-wise masses, activities, radiotoxicities and heat productions in the system at each time step. However, the results are only provided for nuclides specifically chosen by the user. For an experienced reactor physicist the nuclides of interest in a problem may be obvious, but a user with less experience might face serious problems when trying to identify the important nuclides in the vast selection.

To further increase the usability of Serpent, a new intelligent nuclide selection capability for the output of Serpent 2 is developed and implemented in the code. The new capability is designed with especially final disposal, transmutation and other fuel cycle -related analyses kept in mind. The new routine selects the output nuclides on basis of their contributions to a user-chosen property. For example, the user may request results for the 10 nuclides with the highest decay heat production. The new version of Serpent also includes a selection of pre-defined nuclide groups for different purposes involving, for instance, the nuclides with a high migration probability and the minor actinides. Also the nuclide list used in COSI6 is included to provide for easy importing of materials from Serpent to COSI6.

This paper describes the implementation of the new nuclide selection capability in Serpent 2. The applicability of Serpent in inventory calculations is also discussed through practical examples.

Reference

[1] Serpent website, http://montecarlo.vtt.fi