RECENT RESEARCH AND DEVELOPMENT ACTIVITIES ON PARTITIONING AND TRANSMUTATION OF RADIOACTIVE NUCLIDES IN JAPAN

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Abstract

In Japan, R&D activities for partitioning and transmutation (P&T) have been promoted under the OMEGA programme for more than 15 years. These activities were reviewed by the Atomic Energy Commission in Japan in 2000. In accordance with the results of the review, three institutes, the Japan Atomic Energy Research Institute (JAERI), the Japan Nuclear Cycle Development Institute (JNC) and the Central Research Institute of Electric Power Industry (CRIEPI), are continuing the R&D on the P&T technology. This report summarises the recent activities in Japan by these institutes. JAERI is engaging in the R&D on the Double-strata Fuel Cycle concept consisting of the partitioning process of the high-level waste and the dedicated transmutation cycle using the accelerator driven system (ADS) fuelled with the minor actinide (MA) nitride fuel. JNC and CRIEPI are engaging in the R&D on the P&T technology using commercialised fast reactors (FR), where JNC is mainly in charge of the MOX fuel and the aqueous reprocessing, while CRIEPI is mainly in charge of the metallic fuel and the dry reprocessing. The R&D activities on FR are organised under the Feasibility Study on Commercialised Fast Reactor Cycle Systems.

Introduction

In Japan, the spent fuel discharged from nuclear reactors is to be reprocessed, taking account of the low self-sufficient rate of the energy resources. To fulfil the nuclear fuel cycle, the disposal of the high-level radioactive wastes (HLW) discharged from the reprocessing plant should be steadily implemented. In Japan, the legal framework and the implementation entities were established around the year of 2000.

In parallel to such a movement, the Japanese Government has promoted the research and development (R&D) activities for partitioning and transmutation (P&T) of radioactive nuclides under the long-term R&D programme called OMEGA for more than 15 years, aiming at the reduction of the burden of the backend of the nuclear fuel cycle. These activities were reviewed by the Atomic Energy Commission in Japan in 2000. In accordance with the results of this review, three main institutes in Japan are continuing the R&D on the P&T technology. The Japan Atomic Energy Research Institute (JAERI) is engaging in the R&D on the Double-strata Fuel Cycle concept consisting of the partitioning process of the high-level waste discharged from reprocessing plant and the dedicated transmutation process using the accelerator driven system (ADS) fuelled with the minor actinide (MA) nitride fuel. The Japan Nuclear Cycle Development Institute (JNC) and the Central Research Institute of Electric Power Industry (CRIEPI) are engaging in the R&D on the P&T technology using commercialized fast reactors (FR), where JNC is mainly in charge of the MOX fuel and the aqueous reprocessing, and CRIEPI is mainly in charge of the metallic fuel and the dry reprocessing. This report overviews the recent R&D activities by these institutes.

Activities in JAERI

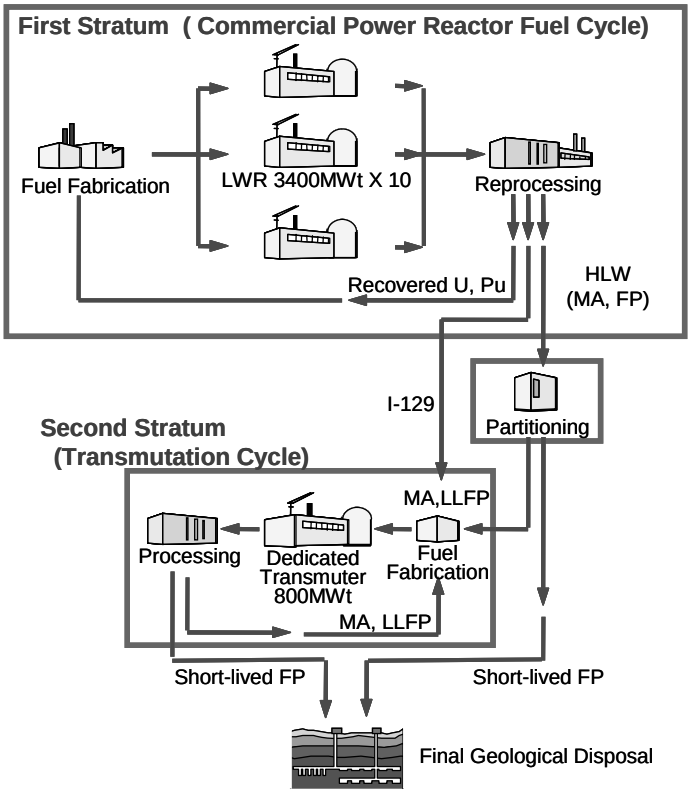
JAERI has proposed the P&T technology based on “Double-strata Fuel Cycle Concept” [1]. Figure 1 shows a schematic outline of this concept. A dedicated transmutation fuel cycle, i.e. the “second stratum”, is established separately from the commercial power generation fuel cycle, i.e. the “first stratum”, where the plutonium is recycled. The HLW exhausted from the reprocessing plant in the first stratum is treated at the partitioning plant and separated into four groups: transuranic elements (TRU), Tc and noble metal, Sr-Cs, and the other elements. The TRU mainly consisting of minor actinides (MA), namely Np, Am and Cm, is the principal object of the transmutation because these elements dominate the potential radio-toxicity in the HLW for a long term later than hundreds years after the reprocessing.

In the transmutation fuel cycle, nuclear fuel mainly consisting of MA is used to enhance the transmutation efficiency. Using such MA fuel, the critical reactor would encounter some difficulties in its safety and controllability aspects. The accelerator-driven subcritical system (ADS) is, therefore, selected as the first candidate of the dedicated transmutation system in the Double-strata Fuel Cycle Concept.

There are, however, several technical issues to be solved for the realization of a large-scale ADS. In JAERI, therefore, various research and development (R&D) activities on the ADS are being conducted in the field of the proton accelerator, lead-bismuth eutectic (LBE or Pb-Bi) and subcritical reactor physics as well as the conceptual design study of a large-scale ADS.

It is also important to evaluate the impact of the P&T technology on the waste disposal concept. The preliminary results of the evaluation are also briefly mentioned afterwards.

Figure 1. **Double-strata fuel cycle concept in JAERI**

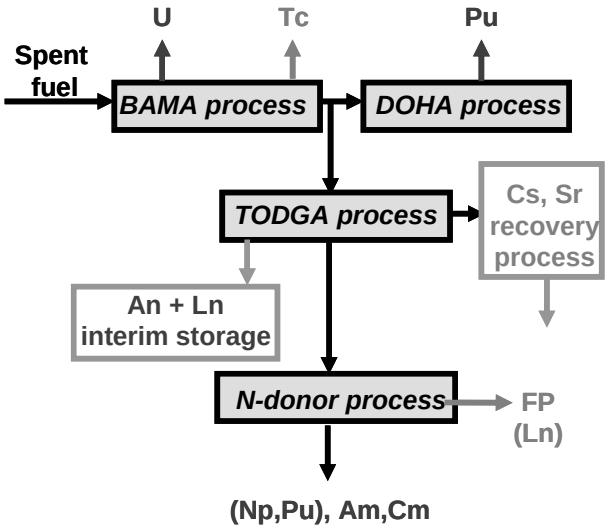


R&D on partitioning process in JAERI

After the establishment of the "4-group Partitioning Process Concept", the basic experimental confirmation was implemented by using condensed high-level radioactive liquid waste [2]. The size of the experiment was about 1/1000 of the expected commercial plant. It was confirmed that Am, Cm, the platinum group, Sr and Cs can be separated as expected.

Although the 4-group Partitioning Process is considered as the reference process in the Double-strata Fuel Cycle Concept, another innovative concept is also being studied to improve the partitioning performance. This concept is called "ARTIST: Amide-based Radio-resources Treatment with Interim Storage of Transuranics" [3], where the object of the process is not the HLW but the spent fuel. The extract agents called BAMA and TODGA are used for uranium separation and all TRU separation, respectively, as shown in Figure 2. The advantages of the process are to use phosphorus-free agents consisting of carbon, hydrogen, oxygen, and nitrogen (CHON principle) to reduce the waste from the process. Basic study on the improvement of the process and the development of new extract agent is under way.

Figure 2. **Concept of ARTIST process**



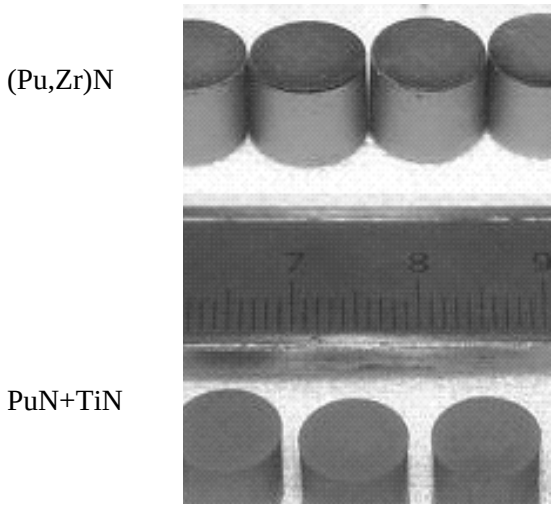
R&D on transmutation fuel cycle in JAERI

JAERI has chosen the MA-nitride fuel as the first candidate for the dedicated transmutation system such as the ADS because of its possible mutual solubility among the actinide mononitrides and its good thermal properties. As the R&D for the fuel fabrication, high-purity nitrides such as NpN, AmN and (Cm,Pu)N were synthesized by the carbothermic reduction method and their material properties have been measured [4]. The solid solution of (Pu,Am,Cm)N was successfully prepared to demonstrate the mutual solubility. MA nitrides with inert matrix such as (Am,Zr)N, (Am,Y)N [5] and (Pu,Am,Cm,Zr)N were also synthesized successfully.

Chemical and thermochemical stability of AmN and (Am,Zr)N was studied in terms of the hydrolysis behaviour at room temperature and the evaporation behaviour at elevated temperatures. In both the cases AmN was stabilized by the formation of solid solution with ZrN. Thermal diffusivity, specific heat capacity and thermal expansion of Am-containing nitrides will be measured.

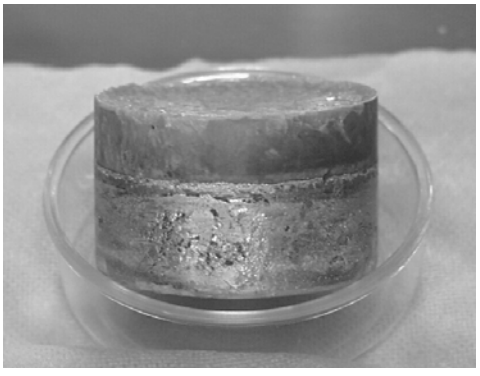
As for the irradiation test, (U,Pu)N fuel was irradiated in the experimental fast test reactor JOYO under the joint research with JNC, and no failure of fuel pins was found [6]. The irradiation test of uranium-free nitride fuels in the Japan Materials Testing Reactor (JMTR) was started in May 2002 [7]. One He-bonded fuel pin incorporating (Pu,Zr)N and PuN+TiN pellets (Figure 3), together with one (U,Pu)N fuel pin as reference, are under irradiation. The irradiation of the fuel pins will be continued till the end of this year, followed by post-irradiation examinations in 2005. Moreover, JAERI will participate in the FUTURIX programme to obtain the information of the irradiation behaviour of MA-bearing nitride fuels in PHENIX reactor in France.

Figure 3. **Appearance of uranium-free nitride fuel pellets**



To reprocess the irradiated MA nitride fuel, the pyrochemical process has been studied in JAERI because it has several advantages over the wet process in treating the dedicated fuel for transmutation including recycling of ^{15}N used in the nitride fuel. In the laboratory scale test, metallic Pu and Np were successfully recovered from non-irradiated PuN and NpN, respectively, by the molten-salt electrorefining technique [8]. Figure 4 shows the Cd cathode after recovery of Pu. A part of these R&D activities are collaborated with CRIEPI. Nitride formation behaviour of Pu-Cd alloy was experimentally investigated. Heating of Pu-Cd alloy at 973 K in N_2 stream resulted in almost complete nitride formation of Pu and distillation of Cd simultaneously.

Figure 4. **Recovery of Pu into liquid Cd cathode**



Regarding the transmutation target for long-lived fission products (LLFP), thermal properties of Tc-Ru were measured [9]. The basic experimental study is under way for the selection of suitable chemical form for the iodine target [10].

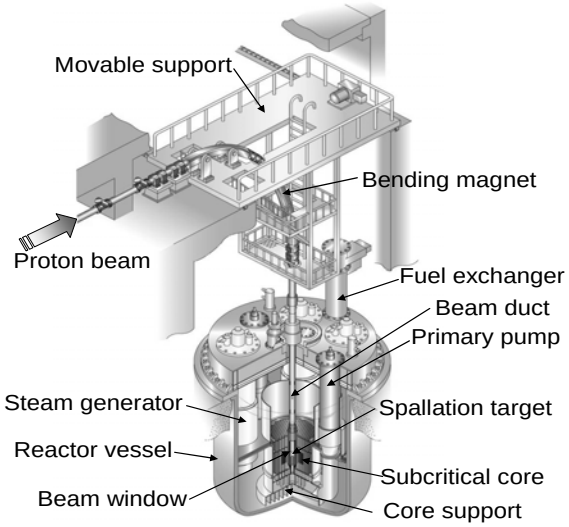
R&D on dedicated transmutation system in JAERI

The R&D of the ADS as the dedicated transmutation system is in progress for three technological areas: the accelerator, the lead-bismuth, and the subcritical reactor. Regarding the accelerator for the ADS, JAERI has chosen the superconducting LINAC as the first candidate. The cryomodule, which is a unit component of the high-energy part of the LINAC, is being fabricated to evaluate its acceleration performance, energy efficiency and stability [11]. In addition to this elemental development, JAERI is constructing a high-intensity proton accelerator under the framework of J-PARC Project (Japan Proton Accelerator Research Complex). The Phase-I of the project will be completed by the fiscal year (FY) of 2007, where a 200 MeV LINAC with 0.33 mA is being constructed as well as two synchrotrons. After the completion of the Phase-I, the LINAC will be upgraded to 400 MeV, which is the original specification of the Phase-I. After this upgrade, the LINAC will be extended up to 600 MeV by SC-LINAC as the Phase-II.

Regarding the lead-bismuth eutectic (LBE), three kinds of R&D activities are mainly under way in JAERI: the material corrosion/erosion test, the thermal-hydraulics test for the beam window and the evaporation test of polonium from LBE [12]. To perform these R&D, JAERI installed three LBE loops and a static corrosion test device. Moreover JAERI collaborates with the Mitsui Engineering and Shipbuilding Co. Ltd. for the corrosion loop test and the thermal-hydraulic loop test, and with JNC for the measurement of the evaporation rate of polonium. As the international collaboration for the LBE target demonstration, JAERI participates in the MEGAPIE Programme in PSI, Switzerland.

The design study of the subcritical reactor is also under way to propose feasible plant concept of 800 MWth ADS which can transmute 250 kg of MA annually [13]. The conceptual view of the subcritical reactor is shown in Figure 5. The LBE is adopted as the spallation target and the core coolant. The feasibility of the plant is being discussed in terms of structural strength of the beam window, the cooling performance of hot-spot pins, and so on [14]. The prediction accuracy of the reactor physics parameters such as the effective multiplication factor and the burn-up reactivity swing is also being discussed. To verify the accuracy of the MA nuclear data, the post irradiation examination (PIE) for the MA samples were implemented and the valuable information was obtained from its analysis [15].

Figure 5. **Conceptual view of 800 MWth LBE-cooled ADS**



As the next step to study the basic characteristics of the ADS and the transmutation technology, JAERI plans to build the Transmutation Experimental Facility (TEF) in the Phase-II of the J-PARC Project [16]. The construction of the TEF is scheduled to start around FY2007. The TEF consists of two buildings: the Transmutation Physics Experimental Facility (TEF-P) and the ADS Target Test Facility (TEF-T). The TEF-P is a zero-power critical facility where a low power proton beam is available to research the reactor physics and the controllability of the ADS. The TEF-T is a material irradiation facility which can accept a maximum 200kW-600 MeV proton beam into the spallation target of LBE.

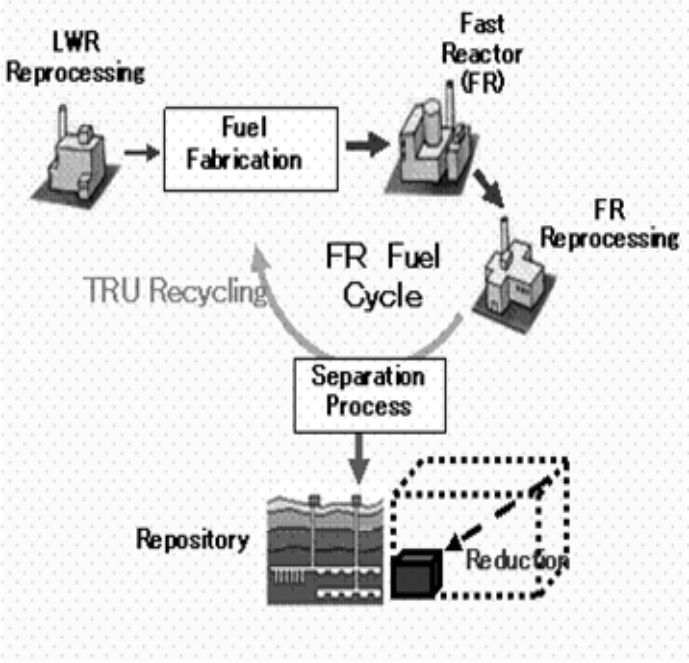
R&D on P&T waste disposal in JAERI

To evaluate the impact of P&T technology on the backend of the nuclear energy utilization, the amounts and forms of the wastes from the partitioning plant and the transmutation fuel cycle were roughly estimated. The results showed that the volume of the vitrified forms can be reduced to about 1/4 by removing Sr-Cs, the platinum group and MA, and they can be disposed twice as densely as the normal case because of their low heat production. The Sr-Cs will be cooled as calcined waste forms, and its disposal method is being investigated. The amount of the wastes from the transmutation fuel cycle will be relatively small because the mass flow of the second stratum in the Double-strata Fuel Cycle Concept will be much smaller, approximately 1/100, than that of the first stratum [17].

Activities in JNC

Research and development of P&T in JNC have been performed in conjunction with the Feasibility Study on Commercialized Fast Reactor Cycle Systems (FS) [18] and as a part of it. Various candidate options, such as sodium-cooled/helium-cooled/lead bismuth-cooled/water-cooled reactors, MOX/metal/nitride fuels, aqueous/oxide electrowinning/metal electrorefining reprocessings, and palletizing/sphere packing/casting fuel fabrications have been studied for the fast reactor fuel cycle system shown in Figure 6 under the FS.

Figure 6. **Advanced fast reactor fuel cycle**



The basic standpoints of JNC for the P&T are as following. All TRU will be treated in the same manner without distinguishing between Pu and MA, although necessity of cooling storage of Cm will be investigated. Homogeneous loading of TRU in fuel assembly will be mainly studied, although some extent of MA target fuel assembly will be studied. FP will be classified into four categories: the transmutation in the form of target assembly after separation (Tc, I), the cooling storage after separation (Sr, Cs), the waste as stable elements after separation (Mo, etc.), and the effective utilization after separation (Ru, Rh, Pd, etc.).

Development goals of P&T technology are the reduction of radiotoxicity, the reduction of high level waste (HLW), and the effective utilization of rare element FP.

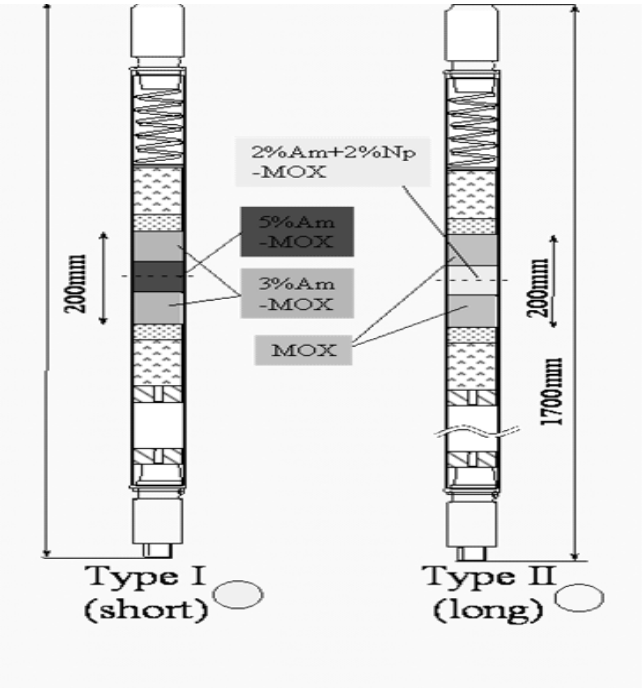
R&D on radiotoxicity reduction in JNC

High Recovery Rate Recycle of TRU

It has been evaluated that more than 99% recovery rate of TRU can be achieved and that there is a possibility to attain the target value of 99.9% by the NEXT (New Extraction System for TRU Recovery) process [19] which is an advanced aqueous reprocessing method developed by JNC. The NEXT process has also been evaluated to have an advantage in waste generation and cost for apparatus [20]. A basic research to develop the extraction system for total recovery of uranium and all transuranics from HLW solutions is in progress as a cooperative study with KRI (Khlopin Radium Institute), Russia [21]. The other hand, a cooperative research with Tokyo Institute of Technology to separate Am and Cm has started.

As for the MA contained MOX fuel development, fabrication experiments of 2%Np-2%Am-MOX pellet and up to 5% Am content MOX pellet have been successfully completed [22] and the irradiation experiment at the fast experimental reactor JOYO with the fuel pin structure shown in Figure 7 is scheduled in 2006.

Figure 7. **Fuel pin structure of MA-MOX irradiation experiment in JOYO**



Critical experiments of Np-loaded core which is a collaboration research with IPPE (Institute of Physics and Power Engineering), Russia, and irradiation experiment of MA (^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm) samples at JOYO have been conducted. Analysis works of both experiments are in progress in order to validate nuclear data and analysis method. It is suggested, through the analyses, that the value of one important parameter for MA composition change, ^{241}Am isometric ratio, would be around 0.85 [23].

P&T of LLFP

Candidate LLFPs to be transmuted in the FS are iodine and technetium. The most significant design limitation for LLFP sub-assembly is found, through the design study of LLFP loaded fast reactor core, to be the temperature of the moderator pins that determines the dissociated hydrogen permeation rate [24]. The out-of-pile tests in order to collect basic data such as sintering characteristics, compatibility with cladding and thermal conductivity have been started for candidate compounds of iodides such as NiI_2, MgI_2, BaI_2, CuI, KI, RbI, and YI_3. Sample irradiation experiment of iodides is scheduled from 2007 at JOYO.

As for the nuclear data, neutron capture cross sections for major FP such as ^{90}Sr, ^{99}Tc, ^{135}Cs, ^{137}Cs have been measured [25, 26] and a collaboration research with ORNL (Oak Ridge National Laboratory) to measure the neutron capture cross sections of ^{93}Zr, ^{99}Tc and ^{107}Pd is in progress [27].

R&D on HLW reduction in JNC

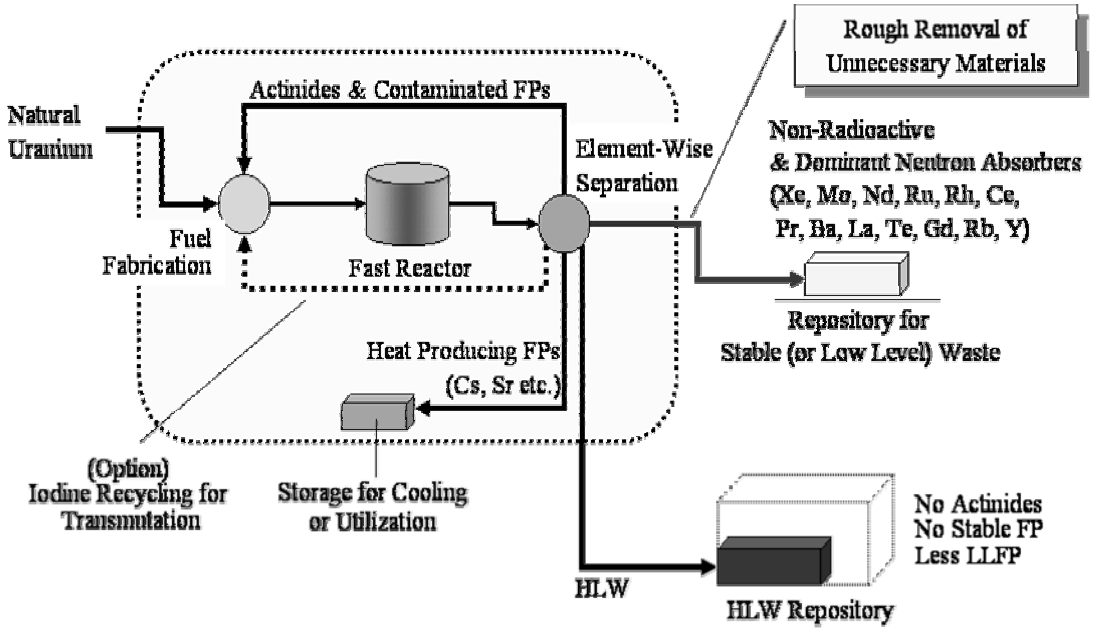
It has been evaluated by the FS that the generated wastes from the various candidate fast reactor fuel cycle options have no significant difference and amount to, in volume, about half of those from the conventional reference system which recycles without MA.

The advanced aqueous reprocessing method developed by JNC aims at to be a salt-free process. Consequently, the vitrified waste produced by the process can be reduced to about 60% of that of the conventional process with the aid of the fact that only small amount of heat generating TRU shifts, as recovery loss, to the HLW in the advanced fast reactor fuel cycle.

Reduction of vitrified waste can also be attained by removing heat generating FP, such as Sr and Cs, as well as Mo which causes soluble phase in glass. Therefore, design study of recovery and temporary storage methods of Cs/Sr and Mo including the economic evaluation have been conducted as a part of the FS.

JNC has proposed a new concept of fast reactor fuel cycle system named “ORIENT-cycle” (Optimization by Removing Impedimental Elements) [28] which aims to minimize HLW by adopting an unconventional recycling scheme based on the idea “rough removal of unnecessary elements” instead of a conventional one “pure recovery of necessary elements” (see Figure 8). Stable FP that amount to about 60 wt% of all FP are identified as one of key “unnecessary elements”. The evaluated result indicates that the ORIENT-cycle might be able to reduce the HLW from aqueous process by about one order of magnitude compared to the conventional process.

Figure 8. **Concept of ORIENT-Cycle**



Cooperative study with University of California, Berkeley, titled "Research on Effective Application of Partitioning and Transmutation Technologies to Geologic Disposal" was started from 2003 in order to evaluate quantitatively the reduction effect of geological repository burden due to the introduction of P&T technologies [29].

R&D on effective utilization of rare element FP in JNC

An attempt to utilize relatively abundant rare metal fission products (RMFP) which amount to about 30 kg content per metric ton of fast reactor spent fuel due to its high burn-up is in progress. An electrolytic extraction method has been studied to separate RMFP (Ru, Rh, Pd, Tc, etc.) from the nuclear spent fuel to utilize them, for example, as catalysts [30]. A small scale experiment shown in Figure 9 indicates that the promising utilization of RMFP will be as FP-catalyst for hydrogen production by water electrolysis.

Figure 9. **Experimental apparatus of electrolysis**

Basic research has started as another attempt of radiochemical approach to utilize Sr and Cs as radioactive source and heat source.

Activities in CRIEPI

CRIEPI has proposed the P&T technology based on the metal fast reactor and its cycle, in which transuranium elements, TRUs, separated from high level liquid waste (HLLW) coming from PUREX reprocessing of spent LWR oxide fuels, should be transmuted [31]. The pyrometallurgical technique is used to separate of TRUs from HLLW. TRUs are mixed in alloy fuels of U-Pu-Zr for transmuting at fast reactors [32]. Spent metal fuels are reprocessed through pyrometallurgical process with electrorefining and reductive extraction, by which minor actinides, Np, Am, and Cm, are recovered together with U and Pu. This fact leads to an advantage on strong proliferation resistance as well as lightening an environmental burden at the waste disposal.

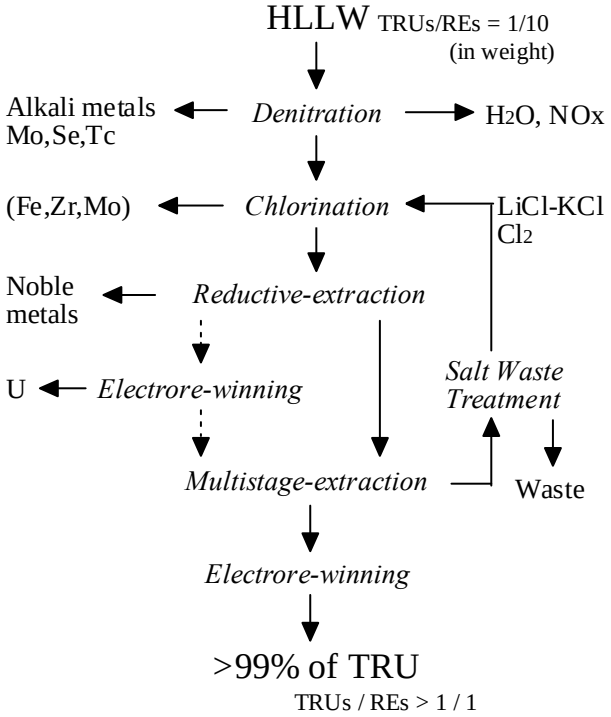
R&D on partitioning process in CRIEPI

Process Description

Figure 10 shows the process flow of separation of U and TRUs from HLLW in which rare earths are existing ca. ten times more than TRUs in weight. The HLLW has to be converted to oxide by heat-treating at 500^{o}C [33]. During this step Tc, Mo, Se and alkali metals are expecting to be separated. The oxides are changed to chlorides by reacting with chlorine gas at 700^{o}C in LiCl-KCl salt bath, in which Fe-, Zr-, and Mo-Clx with high evaporation rate are captured into molten chloride bath trap. The first step of reductive extraction in a system of LiCl-KCl/Cd is applied to separate noble metal elements, and then multistage extraction in a same salt and liquid metal, Cd or Bi, system is possible to separate U and TRUs from rare earths with separation efficiency of more than 99% with attaining the

decontamination factor of over 10. Both reduction steps require lithium metal as reductant, and are operated at 500°C. The advantage of this process is that most of solvents, molten salt, liquid metal and chlorine gas can be recycled after treatment [34].

Figure 10. **Process flow of pyro-partitioning**



Previous and current activity

In the system of molten chloride and liquid metal, the separation of actinides from lanthanides is a key issue from thermodynamic aspect. In CRIEPI, the electrochemical potentials were measured in a cell of M/MCl_n-LiCl-KCl//AgCl-LiCl-KCl/Ag as a function of molar fraction of MCl_n in LiCl-KCl. M denotes an actinide or lanthanide element. Applying Nernst equation on electrochemical potentials measured gives the standard potentials of actinides and lanthanides [35-38]. The potentials promise the precise prediction for separation.

In order to examine the separation efficiency between actinides and lanthanides by reductive extraction, the distribution coefficient of each element was measured in systems of LiCl-KCl/Cd and LiCl-KCl/Bi at 450-500°C, from which the separation factor between actinide and lanthanide is obtained [39, 40]. The LiCl-KCl/Bi system has a higher potential than LiCl-KCl/Cd system for the separation between both elemental groups.

Following the measurements of electrochemical potentials and distribution coefficients, separation tests of actinides have been carried out by reductive extraction. It is assured by multistage reductive extraction experiments with simulated waste that more than 99% of each actinide can be recovered [41]. Through these experiences, it is convinced that the verification with genuine hot material makes an important role for finalizing process flow.

The caisson shown in Figure 11 and Figure 12 has been prepared in hot cell facility by the cooperation of the Institute of Transuranium Elements to use of genuine HLLW produced from reprocessing of irradiated MOX fuels. Prior to the experiment using genuine material, the electrorefining study has been performed to recover Pu from metal fuel for fast reactor fuel cycle programme in CRIEPI. Figure 13 shows the U-Pu-Zr alloy fuel before and after experiment [42].

Figure 11. **External view of the caisson**

Figure 12. **Internal view of the caisson (Electrorefiner operated with manipulator)**

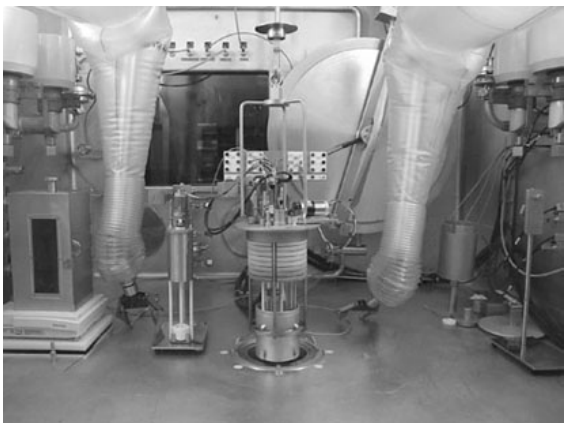
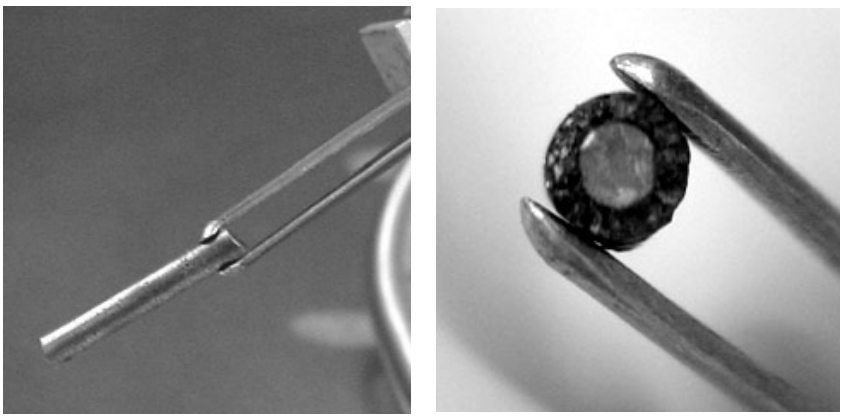


Figure 13. **U-Pu-Zr alloy (left) before and (right) after experiment. U-Pu-Zr alloy remaining after experiment was surrounded by dense layer with Zr metal and salt mixture**



Currently, the programme proceeds to use the genuine HLLW. Some experiments are carrying out to convert HLLW to oxides by evaporating water and decomposing nitrate solution. Chloride conversion and TRUs separation by multistage reductive extraction by use of this oxide are planned in this Japanese FY.

R&D on transmutation in CRIEPI

Characterization of alloy with minor actinides

The actinides are recycled into metal fuel of fast breeder reactors. Phase diagram predicts a low limited solubility of tri-valence species in U-Pu-Zr matrix [43]. U-Pu-Zr alloys added with minor actinides of 2 wt% and 5 wt% each and lanthanides of 2 wt% and 5 wt% each were casted in order to evaluate the miscibility between tri-valence species and U-Pu-Zr. Metallography shows the intermetallic compound of americium, neodymium and cerium distributing uniformly at grain boundaries [44]. The solubility of tri-valence species in U-Pu-Zr is less than 1 wt% even at melting state as observed in EPMA analysis.

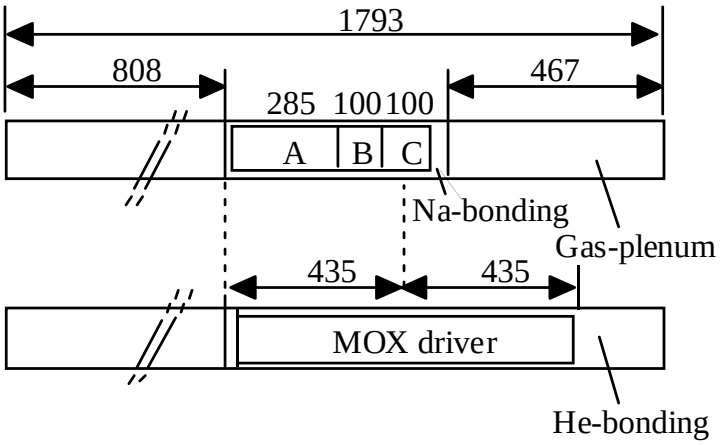
For use of the metal alloy as nuclear fuel, modification of the fundamental properties, such as melting temperature, elastic modulus and redistribution of components under a temperature gradient, has to be clarified on alloys of U-Pu-Zr added with minor actinides and lanthanides [45]. It was observed that the properties of the alloys added with minor actinides and lanthanides up to 5wt% each are approximately same with those of U-Pu-Zr.

The liquefaction temperature by eutectic formation of U-Pu-Zr fuel and stainless steel cladding is expected to be above 600^{o}C, which affects directly the reactor operation temperature. The liquefaction was evaluated from diffusion tests using U-Pu-Zr/Fe couple and the phase diagrams were assessed both by thermodynamic calculation and metallography. The diffusion tests indicate that no melting phase appears under 650^{o}C in a region with less than 25 wt% of plutonium in fuel U-Pu [46, 47]. The thermodynamic calculation of the activity gradient explains the mechanism of liquefaction, namely the diffusion paths through a two-phase region of liquid and U_6Fe, provided that a cladding temperature is greater than 650^{o}C.

Irradiation study of minor actinide containing Alloys

The irradiation study for minor actinide-containing alloy, i.e. U-Pu-Zr-MA(Np, Am, Cm)-RE(Nd, Ce, Gd, Y), is in progress. Nine pins with various concentrations of MA and RE have been fabricated by casting and power metallurgy for high amount of MA and RE. Figure 14 shows figure configuration of irradiation pins for Phenix Fast Reactor. Instead of MOX driver pins, sodium-bonded metal fuel pins shown in the figure were prepared at the Institute of Transuranium Elements. Three kinds of pins, in which 2%MA-2%RE, 5%MA-5%RE and 5%MA in U-Pu-Zr and U-Pu-Zr for comparison are sandwiched with U-Pu-Zr standard alloy, are allocated in a subassembly. Three assemblies with same configuration of pins, METAPHIX -1, -2, -3, are loaded in a core region of PHENIX at the end of 2003, and start the irradiation at the rated output from January 5, 2004. Each assembly will be irradiated to low, medium and high burn-up. METAPHIX 1 discharged from the reactor on August 5, 2004 after a burn up of 2,4 at%. It is also planned that METAPHIX 2 will be unloaded beginning of 2006 with 7 at% and METAPHIX 3 in 2008 with 11 at %. After unloading and cooling, non-destructive examination will be planned for integrity investigation. After NDE, pins will be transported to the Institute for Transuranium Elements, where the destructive examinations, transmutation rate of MA, physical and chemical behaviour of MA in alloys are expected.

Figure 14. **Irradiation test pins for METAPHIX (unit : mm) A, C: U-Pu-Zr reference specimen; B: U-Pu-Zr-MA-RE specimen (diameter of pin = 6.55 mm, cladding = 15/15Ti cw)**



Concluding remarks

The R&D activities for P&T technologies by three Japanese institutes were summarised in this report. JAERI is engaging in the Double-strata Fuel Cycle concept consisting of the partitioning process of the high-level waste and the dedicated transmutation cycle using the accelerator driven system (ADS) fuelled with the minor actinide (MA) nitride fuel. JNC and CRIEPI are engaging in the R&D on the P&T technology using commercialized fast reactors (FR), where JNC is mainly in charge of the MOX fuel and the aqueous reprocessing, and CRIEPI is mainly in charge of the metallic fuel and the dry reprocessing. It should be noted that the unification of JAERI and JNC is scheduled in 2005 so that the major part of the R&D in this report will be continued in the new integrated institute.

Furthermore, in Japan, many activities relating to P&T technologies are also implemented by universities. The Tokyo Institute of Technology (TITech) started a basic study for innovative separation/transmutation systems toward vanishing high-level wastes under a framework of "21st Century Center of Excellence (COE) Programme on Innovative Nuclear Energy Systems for Sustainable Development of the World (COE-INES)". TITech is also conducting the measurement and the evaluation of nuclear data for the MA transmutation by collaborating with JAERI, JNC, Kyoto University, and so on. Kyoto University and the High Energy Accelerator Research Organization (KEK) started the construction of a new-type proton accelerator to couple with an existing critical assembly, though the purpose of this programme is not only for the study of the transmutation but also for the energy production. The Atomic Energy Society of Japan has established the Research Committee on Partitioning and Transmutation Cycle to promote and integrate the various activities mentioned above and to discuss the effect of P&T technology in the context of the waste management.

REFERENCES

[1] T. Mukaiyama, *Importance of Double Strata Fuel Cycle for Minor Actinide Transmutation*, Proceedings 3rd OECD/NEA Information Exchange Meeting on Separation and Transmutation of Actinides and Fission Products, December 12-14, 1994, Cadarache, France, p.30 (1994).

[2] Y. Morita, *et al., A Demonstration Test of 4-Group Partitioning Process with Real High-Level Liquid Waste*, Proceedings International Conference Atalante 2000, October 24-26, 2000, Avignon, France, p.3-37, www.cea.fr/conferences/atalante2000/index.htm (2000).

[3] S. Tachimori, *et al., Recent Progress of Partitioning Process in JAERI: Development of Amide-Based ARTIST Process*, Proceedings 7th OECD/NEA Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, October 14-16, 2002, Jeju, Republic of Korea, CD-ROM (2003).

[4] K. Minato, *et al., Fabrication of Nitride Fuels for Transmutation of Minor Actinides*, Journal of Nuclear Materials, 320, 18 (2003).

[5] A. Itoh, *et al., Fabrication of Americium-Based Nitrides by Carbothermic Reduction Method*, Journal of Nuclear Science and Technology, Supplement 3, 737 (2002).

[6] Y. Arai and T. Ogawa, *Research on Nitride Fuel and Pyrochemical Process for MA Transmutation*, Proceedings 6th OECD/NEA Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, December 11-13, 2000, Madrid, Spain, CD-ROM (2001).

[7] Y. Arai, *et al., Irradiation Test of U-Free Nitride Fuel and Progress of Pyrochemistry in JAERI*, Proceedings 7th OECD/NEA Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, October 14-16, 2002, Jeju, Republic of Korea, CD-ROM (2003).

[8] O. Shirai, *et al., Recovery of Neptunium by Electrolysis of NpN in LiCl-KCl Eutectic Melts*, Journal of Nuclear Science and Technology, 37, 676 (2000).

[9] K. Minato, *et al., Fundamental Properties of Targets for Transmutation of Technetium-99 and Iodine-129*, Proceedings International Conference on Back-End of the Fuel Cycle, Global 2001, September 10-13, 2001, Paris, France, CD-ROM (2001).

[10] Y. Shirasu and K. Minato, *Selection of Chemical Forms of Iodine for Transmutation of ^{129}I*, Journal of Nuclear Materials, 320, 25 (2003).

[11] N. Ouchi, *et al., Development of Superconducting Proton LINAC for AD*S, Proceedings OECD/NEA/NSC International Workshop on the Utilization and Reliability of High Power Proton Accelerators, May 16-19, 2004, Daejeon, Republic of Korea (2004).

[12] H. Oigawa, *et al., Research and Development Program on Accelerator Driven System in JAERI*, Proceedings Global 2003, November 16-20, 2003, New Orleans, USA (2003).

[13] K. Tsujimoto, *et al., Neutronics Design for Lead-Bismuth Cooled Accelerator-Driven System for Transmutation of Minor Actinide*, Journal of Nuclear Science and Technology, 41, 21 (2004).

[14] H. Oigawa, *et al., Design Study Around Beam Window of ADS*, Proceedings OECD/NEA/NSC International Workshop on the Utilization and Reliability of High Power Proton Accelerators, May 16-19, 2004, Daejeon, Republic of Korea (2004).

[15] K. Tsujimoto, *et al., Validation of Minor Actinide Cross Sections by Studying Samples Irradiated for 492 Days at the Dounreay Prototype Fast Reactor – II: Burnup Calculations*, Nuclear Science and Engineering, 144, 129 (2003).

[16] H. Oigawa, *et al., Concept of Transmutation Experimental Facility*, Proceedings OECD/NEA/NSC International Workshop on the Utilization and Reliability of High Power Proton Accelerators, May 16-19, 2004, Daejeon, Republic of Korea (2004).

[17] S. Nakayama, *et al., Partitioning-Transmutation Technology: a Potential for a Future Nuclear Waste Management Option*, Proceedings the RRTD 2nd International Workshop on Nuclear Fuel Cycle, March 24-25, 2003, Aomori, Japan (2003).

[18] Y. Sagayama, *et al., Overall Plan and Progress Situation of the Feasibility Study on Commercialized FR Cycle Systems*, Proceedings Global 2003, November 16-20, 2003, New Orleans, USA (2003).

[19] T. Takata, Y. Koma, K. Sato, *et al., Conceptual Design Study on Advanced Aqueous Reprocessing System for Fast Reactor Fuel Cycle*, Journal of Nuclear Science and Technology, 41, 307 (2004).

[20] Y. Koma, T. Takata and K. Sato, *Design Study on Minor Actinides Recovery for the NEXT Reprocessing*, Proceedings Global 2003, November 16-20, 2003, New Orleans, USA (2003).

[21] M. Ozawa, *et al., Potential of Bifunctional Neutral Phosphoorganic Compounds for Reprocessing of Spent Fuel*, Proceedings International Conference on Back-End of the Fuel Cycle, Global 2001, September 10-13, 2001, Paris, France, CD-ROM (2001).

[22] H. Yoshimoti, *et al., Fabrication Technology for MOX Fuel Containing AmO_2 by an In-Cell Remote Process*, Journal of Nuclear Science and Technology, 41, 1 (2004).

[23] S. Ohki, *Validation of MA Nuclear Data by Sample Irradiation Experiments with the Fast Reactor JOYO*, to be presented ND 2004, Santa Fe, USA (2004).

[24] N. Takaki and T. Mizuno, *Design Study on Sodium-Cooled Fast Reactor Core Loaded with LLFP Transmutation Sub-Assemblies*, Proceedings GENES4/ANP2003, September 15-19, 2003, Kyoto, Japan (2003).

[25] S. Nakamura, *et al., Measurement of the Thermal Neutron Capture Cross Section and the Resonance Integral of the $^{90}Sr(n,\gamma)^{91}Sr$ Reaction*, Journal of Nuclear Science and Technology, 38, 1029 (2001).

[26] H. Harada, *et al., Measurement of Neutron Capture Cross Sections of Long-Lived Fission Products*, Astrophysics, Symmetries, and Applied Physics at Spallation Neutron Sources, p.131 (2002).

[27] K. Furutaka, S. Nakamura, H. Harada, S. Raman and P. Koehler, *Interim Report on Research between Oak Ridge National Laboratory and Japan Nuclear Cycle Development Institute on Neutron-Capture Cross Sections by Long-Lived Fission Product Nuclides*, Document on Collaborative Study, JNC TY8400 2004-001.

[28] N. Takaki, *et al., ORIENT-Cycle: An Evolutional Recycle Concept with Fast Reactor for Minimizing High Level Waste*, Proceedings 7th OECD/NEA Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, October 14-16, 2002, Jeju, Republic of Korea (2003).

[29] J. Ahn and T. Ikegami, *Effective Application of Partitioning and Transmutation Technologies to Geologic Disposal*, in this conference.

[30] M. Ozawa, *Fission Product Recycling as Catalysts for Hydrogen Production by Water Electrolysis*, Proceedings Atalante 2004, June 21-25, Nîmes, France (2004).

[31] T. Inoue and H. Tanaka, *Recycling of Actinides Produced in LWR and FBR Fuel Cycles by Applying Pyrometallurgical Process*, Proceedings of Global 97, Yokohama, Japan, October 5-10, 1997, Vol.1, p.646 (1997).

[32] T. Inoue, *et al., Development of Partitioning and Transmutation Technology for Long-Lived Nuclides*, Nuclear Technology, 93, 206 (1993).

[33] M. Kurata, *et al., Conversion of Simulated High-Level Liquid Waste to Chloride for the Pretreatment of Pyrometallurgical Partitioning Process*, Journal of Nuclear Science and Technology, 37, 682 (2000).

[34] K. Kinoshita, M. Kurata and T. Inoue, *Estimation of Material Balance in Pyrometallurgical Partitioning Process of Transuranium Elements from High-Level Liquid Waste*, Journal of Nuclear Science and Technology, 37, 75 (2000).

[35] Y. Sakamura, *et al., Distribution Behaviour of Plutonium and Americium in LiCl-KCl Eutecti/Liquid Cadmium Systems*, Journal of Alloys and Compounds, 321, 76 (2001).

[36] Y. Sakamura, *et al., Thermodynamic of Neptunium in LiCl-KCl Eutectic / Liquid Bithmuth Systems*, Journal of the Electrochemical Society, 147, 642 (2000).

[37] J. J. Roy, *et al., Thermodynamic Properties of U, Np, Pu and Am in Molten LiCl-KCl Eutectic and Liquid Cadmium*, Journal of the Electrochemical Society, 143, 2487 (1996).

[38] S. P. Fusselman, *et al., Thermodynamic Properties of Rare Earths and Americium in Pyropartitining Process Solvents*, Journal of the Electrochemical Society, 146, 2573 (1999).

[39] M. Kurata, Y, Sakamura and T. Matsui, *Thermodynamic Quantities of Actinides and Rare Earth Elements in Liquid Bismuth and Cadmium*, Journal of Alloys and Compounds, 234, 82 (1996).

[40] M. Kurata, *et al., Distribution Behaviour of Uranium, Neptunium, Rare-Earth Elements and Alkaline-Earth Metals between Molten LiCl-KCl Eutectic Salt and Liquid Cadmium or Bismuth*, Journal of Nuclear Materials, 227, 110 (1995).

[41] K. Kinoshita, *et al., Separation of Uranium and Transuranic Elements from Rare Earth Elements by Means of Multistage Extraction in LiCl-KCl/Bi System*, Journal of Nuclear Science and Technology, 36, 189 (1999).

[42] K. Kinoshita, *et al., Separation of Actinides from Rare Earth Elements by Means of Molten Salt Electrorefining with Anodic Dissolution of U-Pu-Zr Alloy Fuel*, Journal of Physics and Chemistry of Solids, in print.

[43] *Binary alloy phase diagram*, 2nd ed., Ed. T. B. Massalski, ASM International (1990).

[44] T. Inoue, M. Kurata, L. Koch, J-C. Spirlet, C. T. Walker and C. Sari, *Characterization of Fuel Alloys with Minor Actinides*, Transactions of American Nuclear Society, 64, 552 (1991).

[45] M. Kurata, T. Inoue and C. Sari, *Redistribution Behaviour of Various Constituents in U-Pu-Zr Alloy and U-Pu-Zr Alloy Containing Minor Actinides and Rare Earths in a Temperature Gradient*, Journal of Nuclear Materials, 208, 144 (1994).

[46] M. Kurata, K. Nakamura and T. Ogata, *Thermodynamic Evaluation of Quaternary U-Pu-Zr-Fe System for Designing Cladding Temperature of Metallic Fuel Fast Reactor*, Proceedings of 16th IUPAC Conference on Chemical Thermodynamics and of 10th Symposium on Thermodynamics of Nuclear Materials, Halifax, Nova Scotia, Canada, August 6-11, 2000.

[47] K. Nakamura, *et al., Phase Relations in the Quaternary Fe-Pu-U-Zr System*, Journal of Nuclear Materials, 304, 63 (2002).