

**OMEGA PROGRAMME
PARTITIONING AND TRANSMUTATION R&D PROGRAMME OF JAPAN**

1. Overview of OMEGA programme

In 1973, the Japan Atomic Energy Industry Forum published a report entitled “A closed system for radioactivity”. The report pointed out the importance of R&D for P&T of long-lived nuclides as long-term efforts in developing a complete system for radioactive waste management. JAERI started the development of partitioning process for high-level liquid waste and the design study of transmutation system in mid 1970s.

Even after the pessimistic conclusions for P&T drawn by ORNL (1980), IAEA (1982), etc., a small number of groups in Japan and other countries continued basic P&T studies. Based on their studies, the interested Japanese organisations proposed to initiate a major R&D programme on P&T.

In October 1988, Atomic Energy Commission (AEC) of Japan launched a long-term programme for research and development on nuclide partitioning and transmutation technology, called “OMEGA”. The R&D programmes were jointly stimulated by the collaborative efforts of JAERI and JNC. In the public sector, CRIEPI also has been carrying out R&D on this subject.

The programme is conceived as research efforts to assessing the possible benefits of P&T technology for the future generations through the long-term basic R&D, but not to seek a short-term alternative to established or planned fuel cycle back-end policies. The programme is expected to serve to revitalise nuclear R&D in general, and also to attract capable young researchers dedicated to bringing the nuclear option into the 21st century in a healthy state. In addition, advancement of technologies will provide spin-offs for other fields of science and technology.

The programme is to be proceeded in two steps: the phase-I was originally intended to cover a period up to about 1996, and the phase-II to about 2000. In general, the basic studies and tests are to be conducted in the phase-I to evaluate various concepts and to develop required technologies. In the phase II, engineering tests of technologies or demonstration of concepts are planned. After 2000, pilot facilities will be built to demonstrate the P&T technology. The first check and review of the phase-I of the programme is scheduled in late 1998.

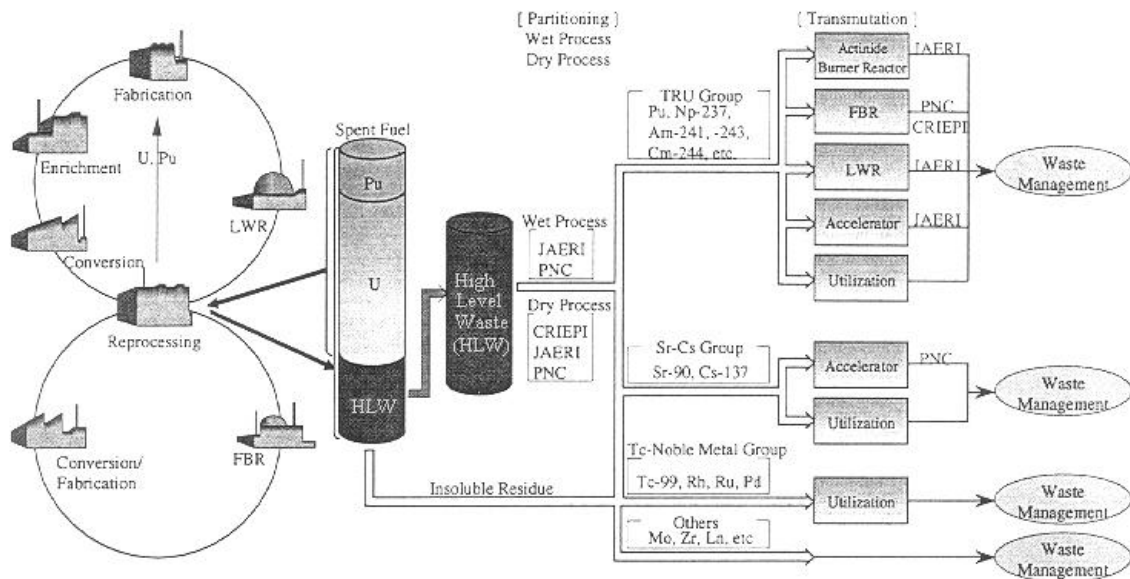
The R&D areas covered by the programme are illustrated in Figure B1. Following items are being studied:

- physical and chemical properties of minor actinides and fission products;
- partitioning of radioactive elements form high-level liquid waste of reprocessing process and recovery of usable metals; and

- transmutation: nuclear and fuel property data of minor actinides, system design studies, reactor fuel and accelerator target development, development of high power accelerator for transmutation.

JAERI has been developing technologies for a dedicated partitioning process and transmutation system based on the double strata fuel cycle concept. JNC has been devoting its major efforts to develop an advanced fuel recycling system with TRUEX process for U, Pu and MA co-extraction and with MOX-FBR for transmutation. CRIEPI has been developing an advanced recycling technology, based on pyroprocess, for metallic-fuelled FBR.

Figure B1. P&T R&D activities under OMEGA programme



2. R&D activities in JAERI

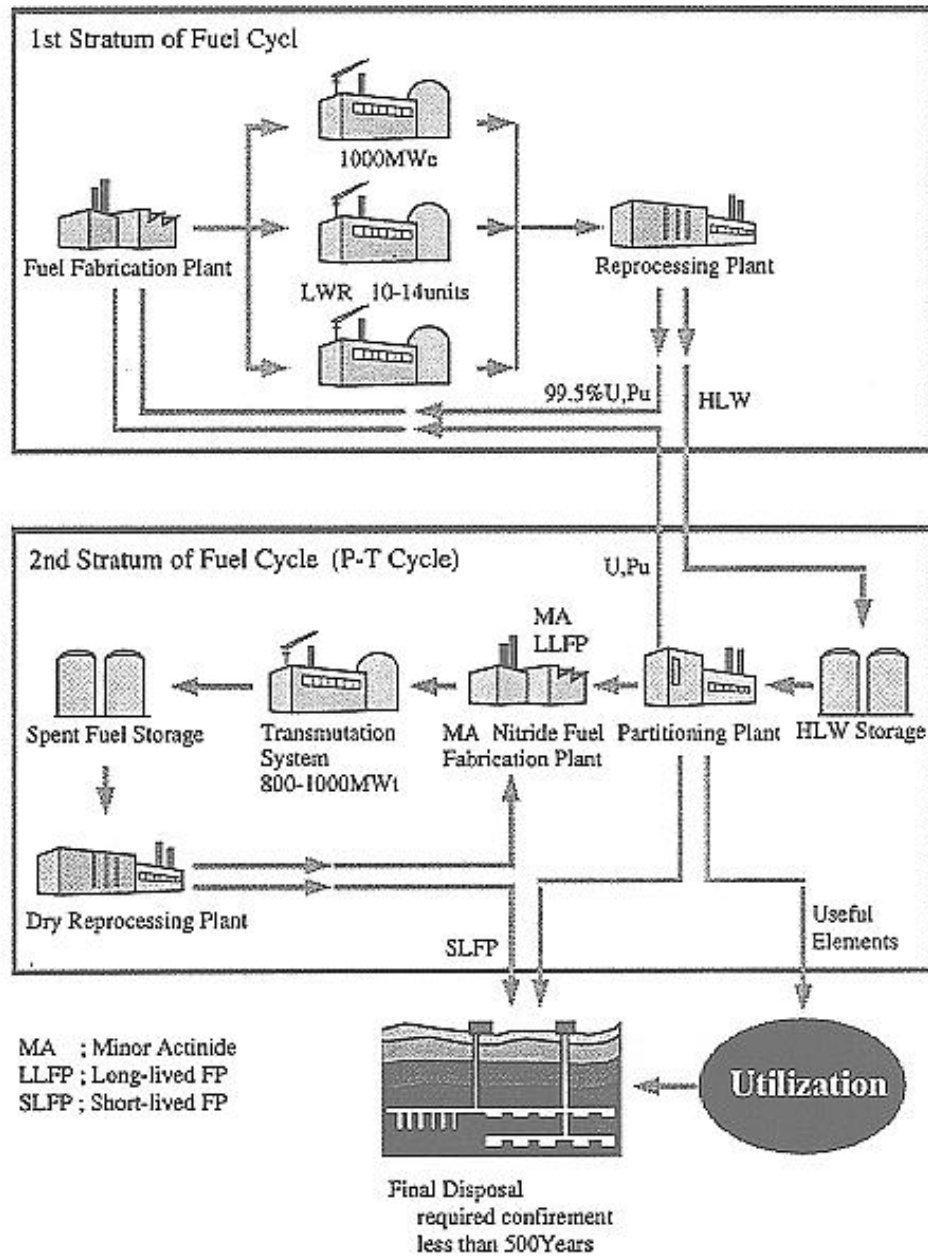
Since the mid 1970s, JAERI has been developing a partitioning process of high-level liquid waste (HLLW) and a concept of a dedicated transmutation system based on the double strata fuel cycle concept (Figure B2). The goal of technological development is the 99.5% reduction in the inventory of MA, ^{99}Tc and ^{129}I which are source terms for long-lasting potential hazard (Figure B3). JAERI's activities cover the following areas: (a) development of the four-group partitioning process, (b) design study of the actinide burner reactor and the accelerator-driven system, (c) development of an intense proton accelerator, (d) development of nitride fuel cycle technologies, and (e) basic research for supporting the development of transmutation systems.

2.1 Double strata fuel cycle [1]

JAERI developed the concept of the double strata fuel cycle, which consists of a commercial fuel cycle and a P&T cycle. A remarkable point is that the latter cycle is independent from the former cycle, i.e., P&T is dedicated for HLW management. Since a P&T cycle handles only the elements contained in the high-level waste solution from commercial reprocessing, the heavy metal throughput

the cycle is about 1/30 of that of the commercial fuel cycle, which will result in compact and economical P&T cycle facilities and in effective transmutation. Introduction of P&T may be easier due to the independence of P&T from the commercial fuel cycle. Troublesome MAs, from the viewpoint of dealing with higher fast-neutron emission and larger α -decay heat, can be confined in the small P&T cycle.

Figure B2. JAERI's concept of Double Strate Fuel Cycle



2.2 Development of four-group partitioning process [2]

A partitioning process was developed for separating elements in HLLW into four groups: TRU, Tc-platinum group metal, Sr-Cs, and others. The process consists of three steps: (a) DIDPA

extraction process separating TRU and lanthanides, (b) separation of Tc-platinum group metal by denitration with formic acid or adsorption with active carbon, and (c) adsorption of Sr and Cs with inorganic ion exchangers (Figure B4). In a laboratory-scale test with simulated HLLW, the recovery of more than 99.95% was demonstrated for all actinides. The four-group partitioning process is being tested with actual HLLW at NUCEF in JAERI.

Figure B3. Reduction of potential radiological toxicity of long-lived nuclides (T1/2 30 years) by transmutation

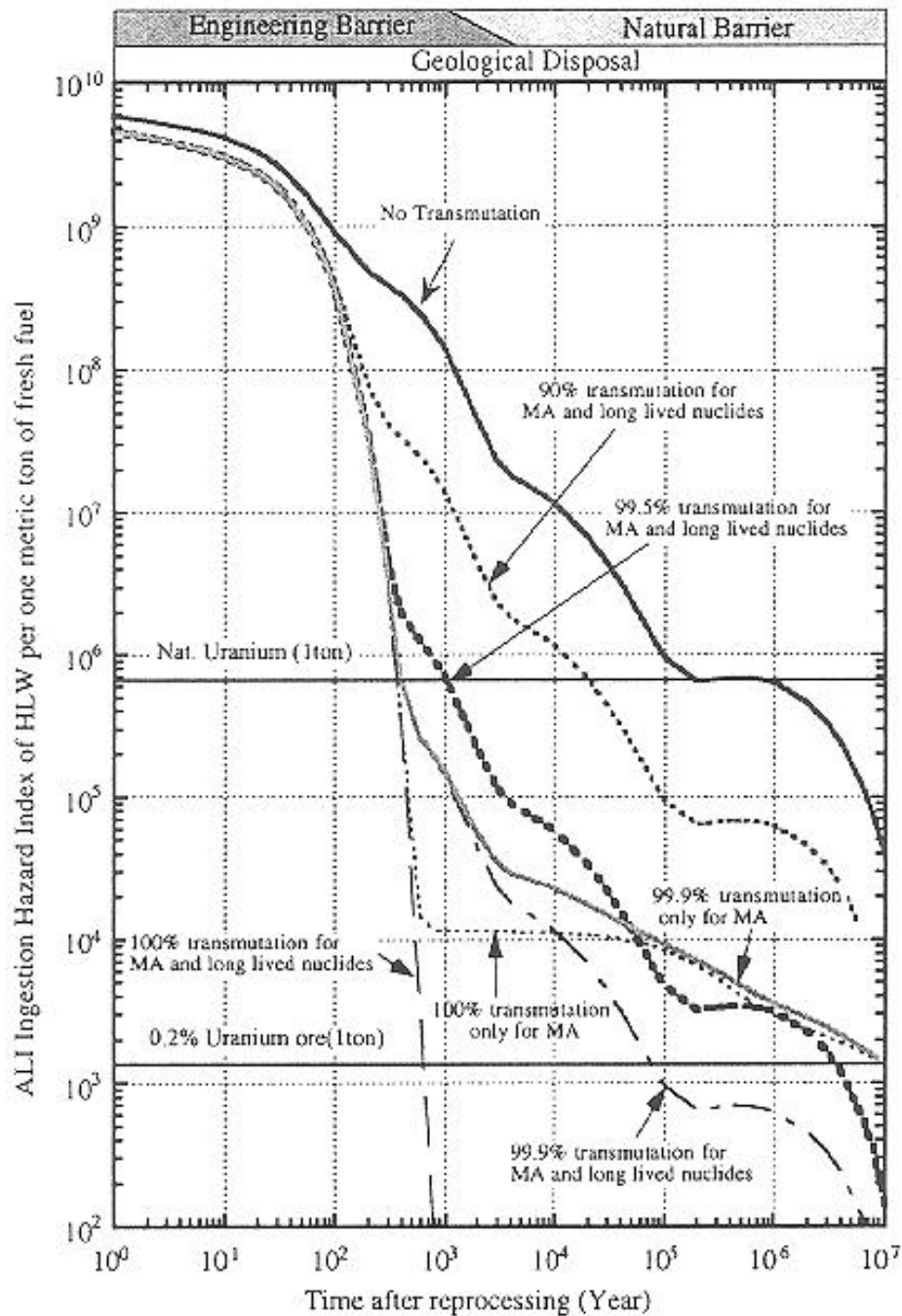
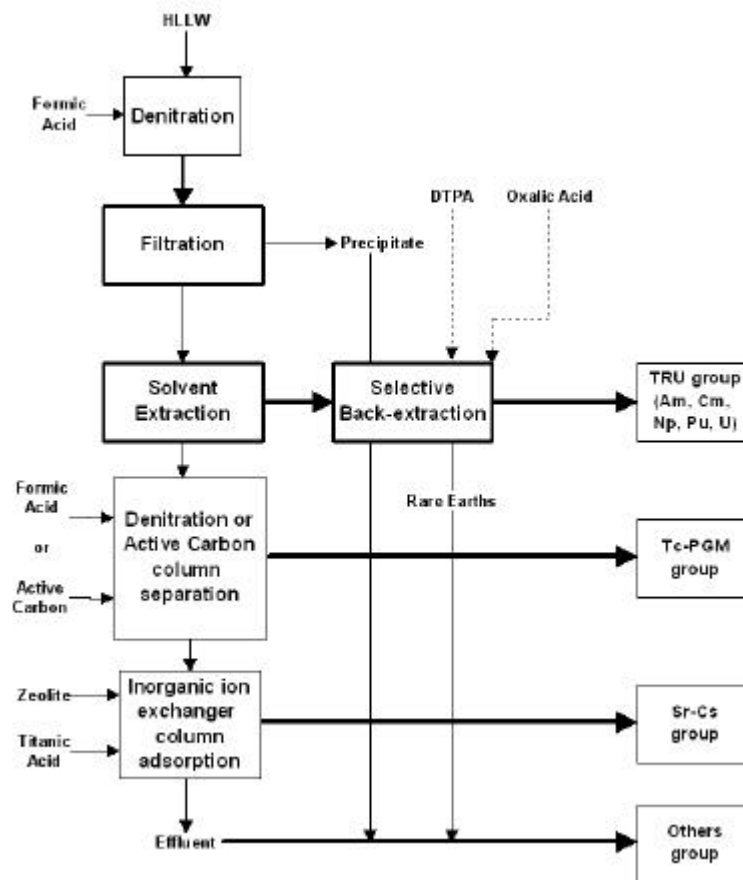


Figure B4. Flow sheet of Four Group Partitioning process



2.3 Design study of the transmutation systems

The neutron physics and ex-core fuel handling process characteristics of MA transmutation were compared among LWR, FBR and a dedicated transmutation system with a very hard neutron spectrum. The generation of heavier MA in LWR and FBR is significantly higher than the dedicated system. The increase of heavier MA needs the reinforced neutron radiation shielding for the fresh fuel handling (manufacturing and transportation) in power reactor fuel cycle. This may cause the increase of electricity generation cost. In the case of a dedicated system, the shielding and the decay heat removal are much severer problem, however, the cost of construction and operation of the dedicated system is not significant because of significantly smaller throughput [3]. From these conclusions, JAERI's design study of the transmutation systems has been devoted to developing the dedicated systems with very hard neutron spectra.

2.3.1 MA burner reactors (ABR: actinide burner reactor) [4]

Two types of ABRs are being studied: a lead-cooled pin fuel ABR (L-ABR) and a helium-cooled particle fuel ABR (P-ABR). Nitride was selected as fuel because of its good thermal property and compatibility with pyrochemical reprocessing. The ABRs have very hard neutron energy spectra with core-averaged energy around 720 keV. The amount of MA fissioned in an ABR is about 250 kg/y.

2.3.2 *Accelerator-driven transmutation system* [5]

Two types of accelerator-driven system are being studied: (a) a solid fuel system with sodium-cooled tungsten target or lead-bismuth target-coolant, and (b) a molten-salt system in which molten chloride salt acts as fuel and target material and also serves as coolant. The solid fuel system (k_{eff} is about 0.95) coupled with a 1.5 GeV - 35 mA proton beam produces 820 MWt and transmutate MA at a rate of approximately 250 kg/y (8% of inventory). The molten-salt system with a 1.5 GeV – 25 mA beam produces 800 MWt. The MA transmutation rate is approximately 250 kg/y (4.6% of inventory).

2.4 *TRU nitride fuel cycle* [6]

The concept of actinide burner cycle for the second stratum (Figure B5) have been proposed, where high densities of the actinides are maintained throughout the whole cycle, and the system volume and envelope are minimised. JAERI is studying the feasibility of nitride fuel and pyrochemical reprocessing. The favourable thermal properties of the nitride fuel make full utilisation of a cold-fuel concept where both the fission-gas release and the swelling of the fuel may be minimised. The requirement on the mechanical strength of the cladding (coating) materials may be eased to have a beneficial effect on realising a harder neutron spectrum.

2.4.1 *Nitride fuel fabrication from actinide salts*

Actinide nitrate solution can be solidified to a ceramic form by an internal gelation method. A droplet of the actinide nitrate solution with a carbon suspension turns into a solid mixture of oxide (hydroxide) and carbon in a form of microspheres. A microwave gelation apparatus has been developed and tested. The oxide+carbon microspheres are converted to the nitride by a carbothermic synthesis. In the particle-fuel concept, the TiN coating consisting of both high-density and low-density layers can be added on the nitride fuel kernel by chemical vapour deposition.

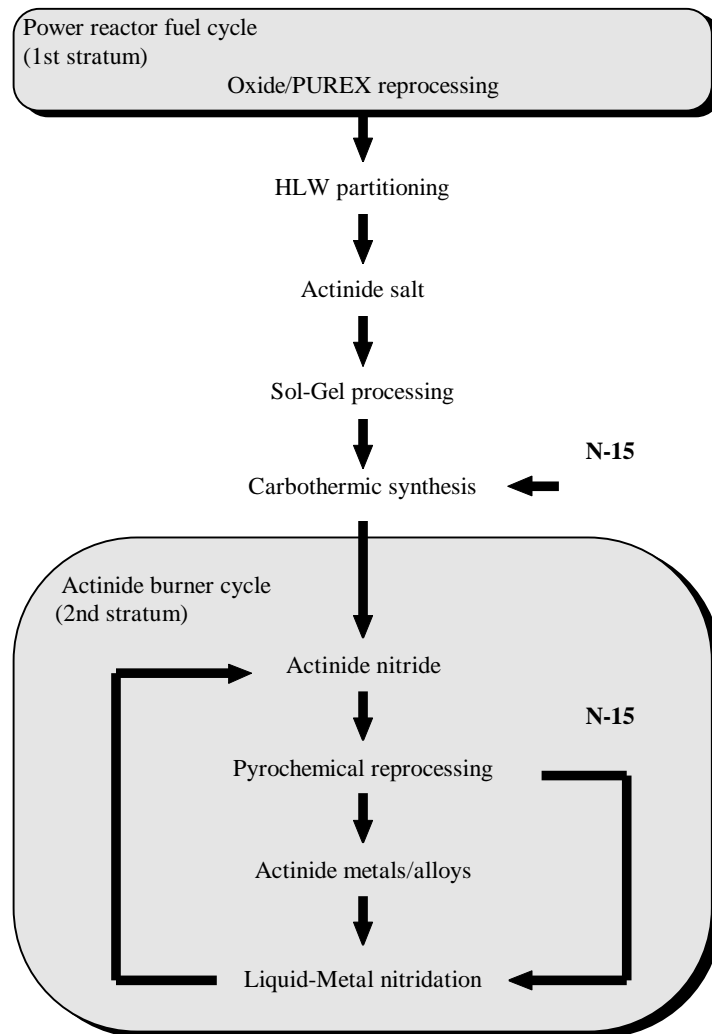
2.4.2 *Electrorefining of nitrides*

In the proposed pyrochemical process, the irradiated nitride fuels are electrorefined in a LiCl-KCl eutectic melt. The recovery of U, Np and Pu metals has been demonstrated in laboratory with the respective nitrides. Conversion of the metal to the nitride can be made by the reaction of N_2 gas with liquid Cd-actinide alloys.

2.5 *Development of a high-intensity super-conducting proton accelerator* [7]

A high-intensity proton linear accelerator with a beam power up to ~8 MW has been proposed for basic science and engineering tests of transmutation systems. Development of an accelerator is carried out as the major development part of JAERI's Neutron Science Project (see below for further details). The main components of the front-end part such as the ion source, RFQ, DTL, and RF source have been constructed. A high brightness (140 mA) hydrogen ion beam has been extracted. The first 2 MeV beam test with the ion source and RFQ in combination of a single unit of RF source was successfully carried out with a peak current of 80 mA at a duty factor of 10%. One superconducting cavity cell was fabricated and tested. The maximum electric field of 44 MV/m was achieved.

Figure B5. Actinide burner cycle with nitride/pyrochemical process



2.6 Basic research supporting the development of transmutation systems

2.6.1 Nuclear data files for transmutation system design study [8]

The JENDL Actinide File contains neutron-induced reaction data of about 90 nuclides from TI-208 to Fm-255. This compilation will be completed by 2000. The JENDL High-Energy File contains the data for high-energy protons and neutrons (up to a few GeV) which are used for the accelerator design and for the accelerator-driven transmutation system study.

2.6.2 Measurement and evaluation of actinide nuclear data [9, 10]

MA nuclear data are measured for fission- and delayed-neutron yields and fission yields in collaboration with ORNL and Texas A&M University. Actinide nuclear data are evaluated using the

integral experiments at FCA. Spallation integral experiments have been carried out at KEK to obtain data and to verify the code system with 500 MeV protons.

2.6.3 *Fuel material thermodynamic database* [11]

A data base of pure substance and solution containing actinides, lanthanides, molten salts, cadmium and nitrogen, related to the actinide burning, is being formed as a supplement to the existing thermodynamic data base.

2.7 *Neutron Science Project* [12]

JAERI has launched the Neutron Science Project with the objective to construct a high-intensity proton accelerator with an beam power of ~8 MW and research facilities for basic science and engineering tests on transmutation systems. Transmutation tests under the project will cover the two steps of accelerator-driven system development. The first step will be a feasibility study of the system concept by using subcritical uranium system at a very low power level. These experiments will check the system operability and MA transmutation capability. The second step will be demonstration tests with a 30-60 MWt experimental reactor. Technical feasibility of spallation target and beam window will be also tested in the second-step.

3. **R&D activities of JNC**

3.1 *Design studies on MA Transmutation in a Fast Reactor* [13-15]

Feasibility studies have been performed to investigate the basic characteristics (transmutation rate, burn-up reactivity, Doppler coefficient, sodium void reactivity, maximum linear heat rate, etc.) of a fast reactor core for MA transmutation. The following items were considered:

- study on loading method of MA in the core (homogeneous, heterogeneous, hybrid, blanket, etc.);
- selection of fuel material for MA transmutation (oxide, inert matrices such as Al₂O₃, CeO₂, etc.);
- study on the maximum tolerable amount of RE nuclides;
- effect of MA recycling on core characteristics and fuel cycle system;
- influence of uncertainties of MA nuclear data, and;
- influence of MA containing fuel on reactor plant and fuel cycle.

The main results of the studies are summarised as follows.

3.1.1 *Study on MA loading method*

The MA transmutation in a fast reactor core has no serious drawbacks in terms of core performance, provided that the homogeneous loading method can be employed with a small ratio of MA

to fuel (~5 wt%). Since a 1 000 MWe-class LWR produces about 26 kg of MA per year, a fast reactor with 5% wt MA loading can transmute the MA mass from six LWRs.

The hybrid MA loading method^{a)} can transmute a large amount of MA without serious drawbacks in terms of core performance. The transmuted mass of MA is about 530 kg/cycle, which is almost 16 times of the mass produced by an LWR with the same power output.

It was found that the hybrid MA loading method, where the Np nuclide is dispersed uniformly in the core and target sub-assemblies containing Am, Cm and rare earth nuclides are loaded into the blanket region, has the potential to achieve the maximum transmutation of MA without special design considerations.

3.1.2 *Study on the permissible RE level in homogeneously-loaded MA fuel*

The homogeneous loading of MA and RE has no serious effects on the reactor core performance, provided that the amounts of MA and rare earths in the fuel are less than 5 and 10 wt% respectively.

3.1.3 *Effect of MA recycling on core characteristics and fuel cycle system.*

The recycling of MA in a fast reactor is feasible from neutronic and thermal-hydraulic points of view. However, during multi-recycling the Np fraction is significantly reduced compared to the unirradiated fuel, and the fraction of Cm is greatly increased because of neutron capture of Am. The accumulation of Cm as a result of the MA recycling will bring about some problems concerning fuel handling and reprocessing, because of the increases in both the decay heat and the neutron emission rate from ²⁴⁴Cm.

3.1.4 *Influence of MA containing fuel on reactor plant and fuel cycle*

Both the decay heat and neutron emission rate of the MA (Np, Am and Cm) loaded fuel are very large in comparison with MOX fuel without MA. The dominant element of these fuel properties is Cm. If it is possible to remove Cm from the MA-loaded fuel, the decay heat value will decrease by one order, and the neutron emission rate by three orders. Since the dominant isotope, ²⁴⁴Cm, has a relatively short half-life of 18 years, there might be another possibility of the fuel cycle, i.e., partitioning of Cm and Am from MA in the reprocessing and storage of Cm and Am for a period.

3.2 ***Measurements of MA and RE cross sections for MA Transmutation in a Fast Reactor***

Fission cross section ratios of minor actinide nuclides (²³⁷Np, ²⁴¹Am and ²⁴³Am) relative to ²³⁵U in the fast neutron energy region have been measured to evaluate the accuracy of MA nuclear data, using a back-to-back (BTB) fission chamber at YAYOI fast neutron source reactor.

a) A combination of the homogeneous and heterogeneous methods: the Np nuclide is loaded in the core region uniformly and a small number of subassemblies containing Am, Cm and RE nuclides are loaded into the blanket region

Making use of BTB fission chambers and a lead slowing-down spectrometer coupled to a 46 MeV electron linear accelerator at Kyoto university, the fission cross sections of ^{237}Np , ^{241}Am , $^{242\text{m}}\text{Am}$ and ^{243}Am have been measured relative to that for $^{235}\text{U}(\text{n},\text{f})$ reaction in the energy range from 0.1 eV to 10 keV. Each of the fission cross sections in the JENDL-3.2 and ENDF/B-VI libraries was compared with the measurement.

As a part of MA nuclear data evaluation, the analysis of irradiated ^{237}Np sample in Joyo has been performed. Additional irradiation test of Np, Am and Cm samples in Joyo was started in August 1994. Measurements of keV-neutron capture cross sections of rare earth nuclides (^{147}Sm , ^{148}Sm , ^{150}Sm , ^{140}Ce , ^{141}Pr , ^{153}Eu , ^{143}Nd and ^{145}Nd) have been performed to evaluate the accuracy of the nuclear data libraries using the 3 MV Pelletron accelerator of the Research Laboratory for Nuclear Reactors at the Tokyo Institute of Technology.

3.3 *Partitioning process based on TRUEX method* [16-18]

As an effort towards establishing partitioning process of actinides in the high level liquid wastes (HLLW), the following R&Ds are being carried out since 1990:

- TRUEX process development for the separation of radioactive elements in HLLW;
- DTPA-TRUEX for separation of MA from Ln ;
- electroredox techniques for back-end solvent extraction processes, and;
- enhancement of Np extraction in PUREX process

The last two R&D items are important supporting techniques for presently assumed advanced reprocessing systems composed of PUREX and TRUEX processes, in order to separate interference (potentially valuable) nuclides like ^{106}Ru and ^{99}Tc prior to solvent extraction cycles as well as to mineralise/minimise radioactive organic waste, and to eliminate Np leakage to HLLW.

Their major achievements and current issues to be solved can be summarised as follows:

3.3.1 *TRUEX process development for separation of actinides and fission products in HLLW*

By several counter-current runs of the TRUEX process using real HAR (highly active raffinates), a process flow sheet capable of selective partitioning of actinides and fission products was newly developed. This flow sheet, named “TRUEX PNC Salt-Free Version”, was characterised by high acidity in the extraction process and by mild salt-free reagents for selective stripping. The process can achieve decontamination factors (DFs) of α -nuclides (including MA) to be $>10^3$, complete scrubbing of Pu and Ru from organic phase ($>>99.5\%$), and improve the recovery yield of MA and Ln (ca. 80-100%) in the aqueous product phase.

The stability of O ϕ D[iB]CMPO in the nitric acid media against γ irradiation (up to the dose of 2.58×10^3 Ci/kg) was sufficient, and thus seemed to be well compatible in each routine partitioning steps. The biological toxicity of O ϕ D[iB]CMPO was as low as TBP. These data were newly obtained by JNC's work.

3.3.2 *DTPA-TRUEX for separation of MA from Ln*

Two counter-current hot runs of DTPA-combined TRUEX process (named SETFICS) achieved separation of MA from MA/Ln mixture in the TRUEX product fraction. As a current result, about 80% of total Ln was removed from ^{241}Am , and the resulted DFs of ^{241}Am against light Ln (La~Nd) were $\gg 20$. Nevertheless, further improvements are still be necessary for heavier Ln (Sm~) separation from MA and for sophistication of the flow sheet, including a method of salt-free waste treatment.

3.3.3 *Electroredox techniques for back-end solvent extraction processes*

By using mediated electrolytic oxidation (MEO), complete destruction of organic wastes from PUREX and TRUEX processes into small inorganic substances was possible. Among various mediators, Ag^{2+} or Co^{3+} ions were found to be proper to destroy $\text{O}\phi\text{D}[\text{iB}]\text{CMPO}$.

By using electrolytic extraction (EE), decontamination of Pd^{2+} , RuNO^{3+} , TcO_4^- (and ReO_4^-) have been investigated. Highly quantitative cathodic deposition was currently obtained for Pd^{2+} and RuNO^{3+} ions from 2.5 M nitric acid by the galvanostatic electrolysis. Deposition of RuNO^{3+} and ReO_4^- was significantly accelerated by the coexistence of Pd^{2+} .

3.3.4 *Enhancement of Np extraction in PUREX process*

In the study of Np extraction by TBP, enhancing extraction of Np was obtained by increasing acidity (e.g. 5.6 M) and temperature (e.g., 100°C) of the feed solution. Addition of chemical redox reagents was not necessary at all.

For establishment of the TRUEX-based partitioning process, the following studies are the most important and must be focused during the next decade:

- hot and engineering-scale counter-current experiments to sophisticate the process flow sheets;
- new ligands for MA/Ln separation, if required separation factor will become greater than hundreds;
- electrochemistry-based “salt-free” techniques towards minimum radioactive waste generation.

3.4 *Transmutation of FP in Accelerator* [19, 20]

3.4.1 *Objective*

It is quite important to pursue a possibility of transmuting long-lived fission products, especially ^{137}Cs and ^{90}Sr , because these nuclides are dominant sources of radioactivity and decay heat during a few hundred years. If we can remarkably reduce these elements, much more radioactive wastes can be stored in a deep geological repository, which will bring considerable benefit of both safety and economy. It is difficult to transmute most of LLFPs in fission reactors, because of their smaller neutron cross sections. Japan Nuclear Cycle Development Institute (JNC) has been studying a possibility to transmute LLFPs, mostly ^{137}Cs and ^{90}Sr using electron accelerator. The basic idea is to apply photo

nuclear reaction, using γ rays produced by an accelerator. The advantages of this method are smaller production of secondary radioactive wastes and broader base of accelerator technology. This work has been conducted as a part of the OMEGA programme in Japan.

3.4.2 *Development of a high power electron linac*

An extremely high power electron linac (e.g. 100 MeV, 1 A) will be required in a future transmutation system, because the yields of the nuclides to be transmuted are high in the spent fuel. It is quite difficult to achieve such a high power in one step. Thus, the beam of 10 MeV and 100 mA (maximum)/20 mA (average) has been selected for the preliminary design study.

The basic and common technical challenges for the high power linac are the suppression of the BBU (Beam Break Up), the removing capability of the heat generated in the linac and also the achievement of high efficiency. To overcome these difficulties, various new ideas such as RF recirculating accelerator structure, advanced RF chopping system for electron beam and 200kV CW electron gun have been adopted to the design.

In addition to the design and analysis, important components such as an accelerator tube and a high power L-band klystron (1.249 GHz RF) were built and many of design concepts which includes an accelerator structure to stabilize high current beam and a heat reducing system for high power RF have been successfully validated. Injector test was conducted in 1996 using a partially-built accelerator and 3 ms pulse width beam of 100 mA peak current was successfully accelerated to the energy of 2.9 MeV. The construction of the linac was completed by March 1997 and tests for beam transport at 10 MeV have been carried out.

3.4.3 *Generation of monochromatic γ ray*

To improve the energy balance of the transmutation by photo nuclear reaction, it is very effective to use monochromatic γ -rays. The energy balance is improved by selecting the energy of the γ -ray beam as that of the resonance peak of the photonuclear reaction.

Various methods have been proposed to generate monochromatic γ -ray, such as supercavity and γ -ray laser. The concept of supercavity is that photons generated by an intense laser system are stored by mirrors of the cavity to which electrons accelerated by high power linac are injected. This makes the probability of Compton scattering three or four orders higher and intense monochromatic γ -ray is expected. Basic experiment has been conducted as a collaboration work with Institute for Laser Technology and JNC.

3.5 *Measurements of cross sections for FP transmutation* [21-26]

JNC has been measuring the thermal neutron capture cross section and the resonance integral of radioactive fission products and have been studying the fine structure in a photonuclear reaction cross section.

To obtain accurate thermal neutron capture cross sections and resonance integrals, the FP target was irradiated by thermal neutrons and the ratio of the activity of neutron capture products to that of FP targets was measured. The thermal neutron capture cross section and resonance integral have

been measured for ^{90}Sr , ^{137}Cs , ^{99}Tc , ^{129}I and ^{135}Cs . For some nuclei, the values of data obtained differs very much from those of previous ones.

For the study of the fine structure in photonuclear cross section, the high energy photon spectrometer with high resolution has been developed. The Monte Carlo simulation showed that the fine structure became observable with an energy resolution of 0.1% by taking advantage of the spectrometer. This study is expected to supply useful data for nuclear transmutation studies using monochromatic photons.

3.6 *Development of Advanced Fuel Recycle System*

JNC started the development of advanced fuel recycle system for a fast reactor breeder reactor pursuing effective waste management, improvement of economy, and enhancement of proliferation resistance. This system is mainly featured by recycling of minor actinides.

Apparently recycling minor actinides in aqueous process needs more equipment, in addition to the PUREX process, which requires somewhat more capital cost. This fact shows that the economical improvement of the advanced fuel recycle system must be reinforced to realise minor actinides recycle.

In this circumstance, JNC proposes a drastically-simplified PUREX process concept, which has good economical performance enough for application of minor actinides recovery. This concept mainly consists of single cycle U/Pu co-extraction process with rather low decontamination factors around 10^3 , which is allowed by the low neutronic sensitivity of a fast breeder reactor to its fuel material impurities. Such simplified extraction process can drastically reduce number and volume of contractors, vessels, waste treatment components, and all auxiliary equipment.

Also there are some supplemental process options to achieve further reduction of equipment. One of the effective options is to apply crystallisation technique to the dissolver solution for the purpose of separating excessive uranium for fabrication of new core fuel before feeding to extraction process.

4. R&D activities in CRIEPI

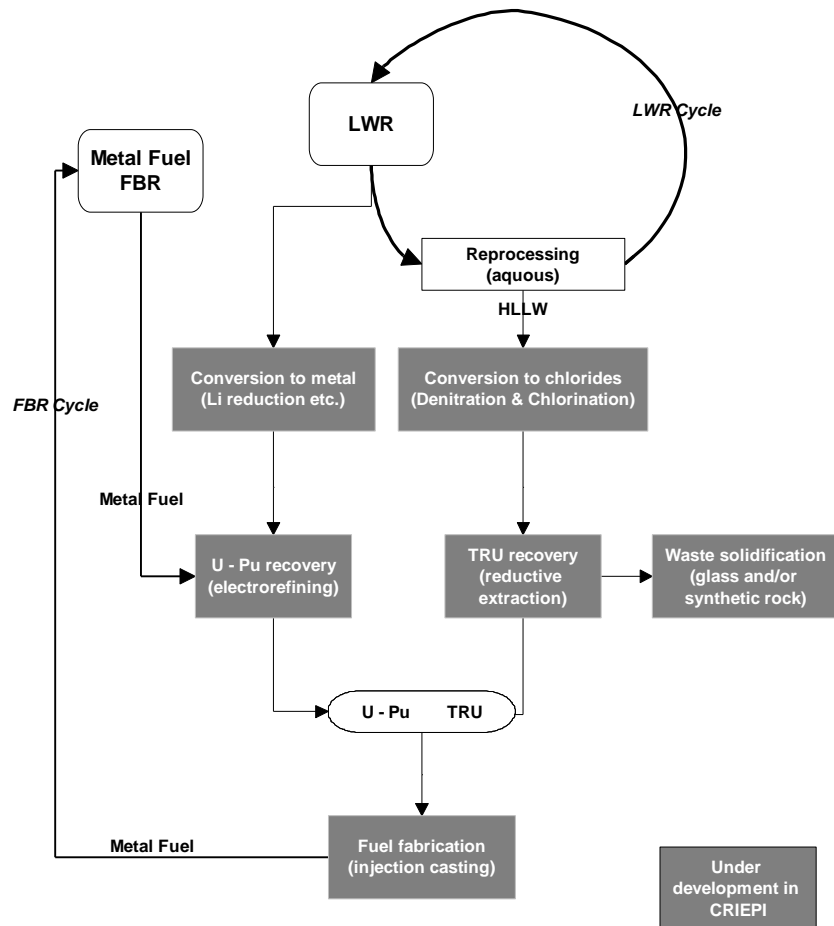
4.1 *Nuclear fuel cycle recycling transuranium elements using pyroprocess and metal-fuelled FBR* [27]

In this scenario, transuranium elements (TRUs) in high level liquid waste and related wastes are separated by pyrochemical method and are transformed to shorter-lived nuclides by burning them in a metal-fuelled FBR. The advantage of this process is to be applied not only to high level liquid waste but also to undissolved residue and concentrates, and solvent scrubbing waste. CRIEPI proposes an actinide recycling system, which contains the programme on pyroprocessing of oxide fuel from LWR and metal fuel from FBR as well as the programme on partitioning and transmutation of TRUs from high level liquid, whose process diagram is shown in Figure B6. Oxide and metal fuels with various burn-up can be reprocessed in the same stream directly by electrorefining to recover uranium and plutonium and by reductive extraction for transuranium elements. Prior to applying electrorefining to oxide fuels, reducing to metals by reductive reagent, such as lithium, has to be conducted.

4.2 *Separation of actinides by pyroprocess from high level liquid waste*

The main separation devices consist of denitration, chlorination and reductive extraction [28,29]. On the pre-treatment process such as denitration and chlorination, technical feasibility was assured by the experimental study, with the results of which the preliminary design study was carried out.

Figure B6. Recycling system for actinides by using pyrochemical process



4.2.1 Measurements of basic data for pyrochemical separation [30, 31]

The distribution coefficients of actinides and rare earths were measured in systems of LiCl-KCl/Cd and LiCl-KCl/Bi with lithium metal reductant. The separation factors to be obtained by the reductive extraction were evaluated.

4.2.2 Study on process flow sheet

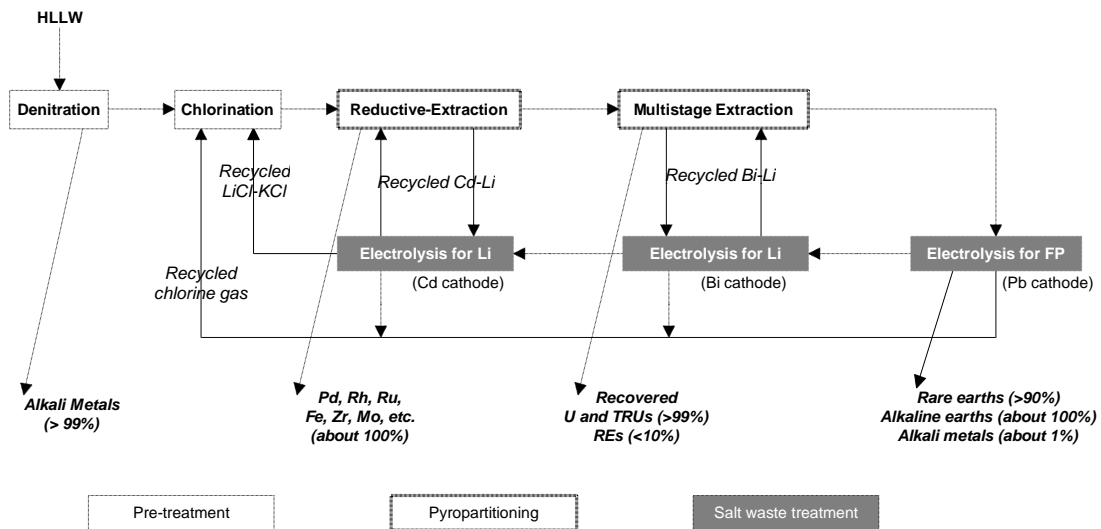
More than 99% extraction of each actinide from salt phase was attained experimentally by multistage reductive extraction. The good separation between TRUs and rare earths is also obtained by applying multistage reductive extraction either in systems of LiCl-KCl/Cd and LiCl-KCl/Bi. The latter system gives better separation with small number of counter-current extraction stages.

4.2.3 Treatment of salt waste [32]

The high-level radioactive waste containing the salt after removing actinides have to be stabilised for disposal. The vitrification process was developed and the flow diagram was proposed. Prior to vitrification, the salts are electrolysed by using liquid lead cathode and the radioactive elements in the lead cathode is oxidised for their separation as oxide slug. This method is expected to produce less secondary wastes because most of solvents, such as LiCl-KCl eutectic salt and lead, are recycled for reuse. The chlorine gas produced by electrolysis is also recycled to chlorination step.

The process flow sheet proposed is presented in Figure B7.

Figure B7. Flow sheet for pyrometallurgical partitioning process



4.3 Transmutation of actinides by metal-fuelled FBR

In this scenario, minor actinides are mixed uniformly in U-Pu-Zr metallic fuel. Following activities are going on in order to evaluate technological feasibility from the points of neutronic characteristics and fuel behaviour.

4.3.1 Analysis of transmutation in metal-fuelled FBR and recycling [33]

The burn-up calculation code, CITATION-TRU, was developed to analyse the transmutation rate of TRUs in FBR. The material flow of actinides in FBR cycle at the equilibrium, when the fuel contains 5 wt% of MA together with some contribution from LWR cycle, suggests that the amount of MA produced in 5 to 6 LWR plants with a capacity of 1 000 MWe in a year can be transformed to other elements in one unit of FBR with a same capacity, as shown in Figure B8.

4.3.2 Core analysis with neutronic safety parameters [34, 35]

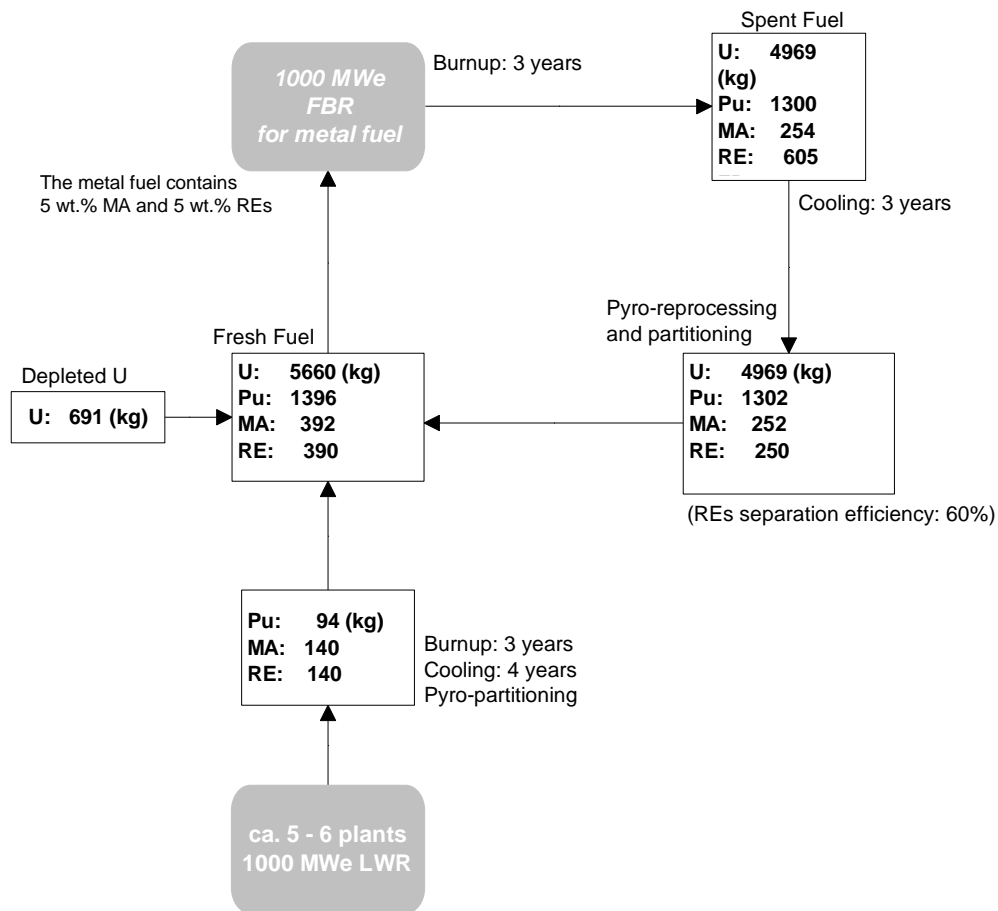
The analysis by CITATION-TRU code showed that the metal-fuelled FBR has safety features unless the content of MA in fuels is more than 5 wt%. It is shown that the reactivity coefficient of

thermal expansion increases slightly with a charge of MA and rare earths, but Doppler coefficient decrease with MA and rare earth loading.

4.3.3 Allowable amount of MA and rare earths in U-Pu-Zr fuel [36]

The miscibility by mixing MA and rare earths in U-Pu-Zr has to be evaluated by observing microstructures. The morphology of U-Pu-Zr alloy contained up to ca. 5 wt% of MA, isotopic composition of which is similar to those discharged from LWR, and of rare earths shows the uniform distribution of rare earth-amerium phase in the matrix, especially along grain boundaries.

Figure B8. **Equilibrium recycle of MA from LWRs in a FBR for metal fuel**



4.3.4 Fuel properties of U-Pu-Zr with MA and rare earths [37, 38]

The characterisation study of alloys with MA was carried out in order to determine the applicability of a metal fuel for FBR. The alloys with 2 and 5 wt% of MA and the alloys containing same amounts of the rare earths were prepared for the examination of the melting temperature, thermal conductivity, elongation and chemical interaction between cladding and alloys. The high evaporation rate of americium was observed above 850°C.

4.3.5 Irradiation study of fuels

The irradiation study is scheduled for alloys containing MA and/or rare earths. The contents of MA and/or rare earths in U-Pu-Zr are 2 or 5 wt% in each. The burn-ups of the fuel alloys waiting for irradiation is expected to be ca. 1.5, 6.0 and >10 at% in a FBR.

REFERENCES

- [1] Murata, H. and Kuroi, H., "A proposed concept for actinide-waste transmutation", in *Nuclear Technologies in Sustainable Energy System* (selected papers from an IIASA Workshop) ed. G.S. Bauer and A. McDonald (Springer-Verlag, 1983)
Murata, T. and T. Mukaiyama, T., "Fission Reactor Studies in View of Reactor Waste Programmes", *Atomkernenergi-Kerntech.*, 45 (1983).
- [2] Kubota, M. and Morita, Y., "Preliminary Assessment on Four Group Partitioning Process Development in JAERI", *Proc. Int. Conf. on Future Nuclear Systems, Global '97*, Vol.1, p. 458 (Yokohama, 1997).
- [3] Mukaiyama, T. et al., "Importance of Double Strata Fuel Cycle for Minor Actinide Transmutation", *Proc. OECD/NEA 3rd Information Exchange Meeting on P&T*, Cadarache (1994).
- [4] Mukaiyama, T., Ogawa, T. and Gunji, Y., "Minor Actinide Burner Reactor and Influence of Transmutation on Fuel Cycle Facilities", *Proc. Technical Committee Meeting on Safety and Environmental Aspects of Partitioning and Transmutation (1993)*, p. 105 (IAEA-TECDOC-783, 1995).
- [5] Takizuka, T., Nishida, T. and Sasa, T., "Conceptual Design Study of Accelerator-Driven Systems for Nuclear Waste Transmutation", *Proc. 2nd Int. Conf. on Accelerator-Driven Transmutation Technologies and Applications*, Vol. 1, p. 179 (Kalmar, 1996).
Tsujiimoto, K., et al., "Conceptual Study of the Lead-Bismuth Cooled Accelerator Driven Transmutation System", to be presented in *2nd ANS Topical Meeting on Nuclear Applications of Accelerator Technology (AccApp'98)*, September 20-23, 1998, Gatlinburg, USA.
- [6] Ogawa, T., Akabori, S., Suzuki, Y., Kobayashi, F., Osugi, T. and Mukaiyama, T., "Nitride Fuel Cycles on Pyrochemistry", *Proc. Int. Conf. on Future Nuclear Systems, Global '97*, Vol.2, p. 812 (Yokohama, 1997).
- [7] Mizumoto, M., Kusano, J., Hasegawa, K., Ito, N., Oguri, H., Tsuchi, Y., Mukugi, K. and Ino, H., "Development of a High Intensity Proton Accelerator", *Proc. 2nd Int. Conf. on Accelerator-Driven Transmutation Technologies and Applications*, Vol. 2, p. 994 (Kalmar, 1996).
- [8] Nakagawa, T., Chiba, S., Fukahori, T., Konshin, Y. and Kikuchi, Y., "Nuclear Data for the OMEGA Project", *Proc. 3rd OECD/NEA Int. Information Exchange Mtg. on Actinide and Fission Product Partitioning and Transmutation*, p. 371 (Cadarache, 1994).
- [9] Oigawa, H., Shinohara, N., Mukaiyama, T., Saleh, H.H., Parish, T.A., Miller, W.H. and Raman, S., "Recent Measurements of Fission Neutron Yield Data of Minor Actinides", *Proc.*

- 4th OECD/NEA Int. Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation*, p.341 (Mito, 1996).
- [10] Takada, H., Meigo, S., Sasa, T., Nishida, T., Takizuka, T., Ishibashi, K. and Nakamoto, T., “Present Status of Integral Spallation Experiments in JAERI”, *Accelerator driven systems: Energy generation and transmutation of nuclear waste - Status Report*, p. 117 (IAEA-TECDOC-985, 1997).
- [11] Arai, Y., Nakajima, K. and Suzuki, Y., “Properties of Neptunium-Plutonium Mixed Nitride Solid Solutions”, *Proc. 4th OECD/NEA Int. Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation*, p. 347 (Mito, 1996).
- [12] Mukaiyama, T., Oyama, Y., Mizumoto, M., Takizuka, T. and Hino, R., “Neutron Science Project at JAERI”, *Proc. Topical Meeting on Nuclear Applications of Accelerator Technology*, p.398 (Albuquerque, 1997).
- [13] Yamaoka, M., Ishikawa, M., and Wakabayashi, T., “Feasibility Study of TRU Transmutation by LMFBRs”, *Proc. Int. Conf. on Fast Reactors and Related Fuel Cycles (FR’91)*, Vol. IV, Oct., 28-Nov. 1, Kyoto (1991).
- [14] Wakabayashi, T. and Ikegami, T., “Characteristics of an LMFBR Core Loaded with Minor Actinide and Rare Earth Containing Fuels”, *Proc. Int. Conf. on Future Nuclear Systems: Emerging Fuel Cycles and Waste Disposal Options (Global’93)*, Vol. 1, Sep. 12-17, Seattle (1993).
- [15] Wakabayashi, T., Ohki, S., Ikegami, T., “Feasibility studies of an optimized fast reactor core for MA and FP transmutation”, *Proc. Int. Conf. on Evaluation of Emerging Nuclear Fuel Cycle Systems (Global’95)*, Vol. 1, Sep.11-14, Versailles, France (1995).
- [16] Ozawa, M., et al., “Enhancing Actinides Separation by Consolidated PUREX and TRUEX Processes Intensified by Salt-free Requisite”, *Proc. Int. Conf. on Evaluation of Emerging Nuclear Fuel Cycle Systems (Global’95)*, Vol. 11, 585-594, Sep. 11-14, Versailles, France (1995).
- [17] Koma, Y., Koyama, T. and Tanaka, Y., “The Concept of Separation Process for Recovery of Americium and Curium”, *Proceedings of Global’97*, Vol.1 598-603, October 5- 10, (1997).
- [18] Ozawa, M., Tanaka, Y., Oohara, C. and Tamura, H., “Back-end Electrochemical Procedures for Waste Minimization in Advanced Actinide Separation System”, *Proceedings of Global’97*, Vol.2 1232- 1237, October 5- 10, (1997).
- [19] Emoto, T., et al., “PNC approach for long-lived fission products transmutation with an electron accelerator”, *Proceedings of Global’97*, October 5- 10, (1997).
- [20] Yamazaki, Y., et al., “Gamma-ray production for applications in nuclear energy”, *Proceedings of ANS Winter Meeting*, November 16-20, (1997).
- [21] Sekine, T., et al., “Measurement of Thermal Neutron Cross Section and Resonance Integral of the $^{137}\text{Cs}(n, \gamma)^{138}\text{Cs}$ Reaction”, *J. Nucl. Sci. Technol.*, 30, 1099 (1993).

- [22] Harada, H., et al., “Measurement of Thermal Neutron Cross Section of the $^{90}\text{Sr}(n,g)^{91}\text{Sr}$ Reaction”, *J. Nucl. Sci. Technol.*, 31, 17 (1994).
- [23] Harada, H., et al., “Measurement of Thermal Neutron Cross Section and Resonance Integral of the $^{99}\text{Tc}(n,\gamma)^{100}\text{Tc}$ Reaction”, *J. Nucl. Sci. Technol.*, 32, 395(1995).
- [24] Nakamura, S., et al., “Measurement of Thermal Neutron Cross Section and Resonance Integral of the $^{129}\text{I}(n,\gamma)^{130}\text{I}$ Reaction”, *J. Nucl. Sci. Technol.*, 33, 283 (1996).
- [25] Katoh, T., et al., “Measurement of Thermal Neutron Cross Section and Resonance Integral of the $^{135}\text{Cs}(n,\gamma)^{136}\text{Cs}$ Reaction”, *J. Nucl. Sci. Technol.*, 34, 431 (1997).
- [26] Harada, H., et al., “High-Resolution Measurement of Fine Structure in the Photoabsorption Cross Section of ^{18}O ”, *Phys. Rev. Lett.*, 80, 33 (1998).
- [27] Inoue, T. and Tanaka, H., *Proc. on Future Nuclear Systems (Global'97)*, pp.646-652, Oct.5-10, 1997, Yokohama, Japan, (1997).
- [28] Inoue, T., Sakata, M., Miyashiro, H., Matsumura, T., Sasahara, A. and Yoshiki, N., *Nucl. Technol.*, **69** 20, 206 (1991).
- [29] Inoue, T., Sakata, M., Kurata, M., Sakamura, Y., Hijikata, T. and Kinoshita, K., IAEA-TECDOC-783, P.137 (1995).
- [30] Sakamura, Y., Inoue, T., Storvick, T.S. and Grantham, L.F., *Int. Conf. on Evaluation of Emerging Nuclear Fuel Cycle Systems (Global'95)*, Vol.2, p. 1185, Versailles, France, Sep.11-14, 1995.
- [31] Kinoshita, K., Inoue, T., Fusselman, S.P., Grimmett, D.L., Roy, J.J., Gay, R.L., Krueger, C.L., Nabelek, C.R. and Storvick, T.S. to be submitted in *J. Nucl. Sci. Technol.*
- [32] Nabeshima, M., Shimizu, T., Sakamura, Y., Sakata, M. and Inoue, T., *J. Nucl. Sci. Technol.* 33, 3, 245, (1996).
- [33] Sasahara, A. and Matsumura, T., *Int. Conf. on the physics of reactors*, Marseilles, France, April 23-26, 1990.
- [34] Sasahara, A., Matsumura, T. and Yokoo, T., *Int. Conf. on fast reactors and related fuel cycles*, P5-13, Oct.28-Nov. 1, Kyoto, Japan, 1991.
- [35] Yokoo, T., Sasahara, A., Inoue T., Kang, J. and Suzuki, A., *Nucl. Technol.* 116, 173, (1996).
- [36] Inoue, T., Kurata, M., Koch, L., Spirlet, J.P., Walker, C.T. and Sari, C., *Trans. Am. Nucl. Soc.*, 6420, 552 (1991).
- [37] Sari, C., Walker, C.T., Kurata, M. and Inoue, T., *J. Nucl. Mater.*, 208, 201 (1994).
- [38] Kurata, M., Inoue, T. and Sari, C., *J. Nucl. Mater.*, 208, 144 (1994).