

**RESEARCH AND DEVELOPMENT OF TECHNOLOGIES FOR PARTITIONING  
AND TRANSMUTATION OF LONG-LIVED NUCLIDES IN JAPAN  
– STATUS AND EVALUATION –**

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## 1. Current activities for radioactive waste management

Measures to treat and dispose of radioactive waste are one of the most important issues in the development and application of nuclear energy. The Atomic Energy Commission (AEC) has carefully considered how to classify radioactive waste properly and how to dispose of it according to these classifications.

Japanese basic policy regarding disposal of high-level radioactive waste (HLW) is to solidify it into stabilized form, to store it for 30-50 years to be cooled, and to dispose of it deep to the underground (geological disposal).

In April 1997, the Advisory Committee on Nuclear Fuel Cycle Back-end Policy, AEC, laid down the guidelines on future research and development of the disposal of HLW. In accordance with the report, the Japan Nuclear Cycle Development Institute (JNC) released the report on the outcome of R&D activities to elucidate technological reliability of the geological disposal, and to provide technical ground for selecting repository sites and for establishing safety requirement in the form of the second progress report in December 1999, with the co-operation of related institutions, such as the Japan Atomic Energy Research Institute (JAERI), the Central Research Institute (CRIEPI), the Geological Survey of Japan (GSJ), the National Research Institute for Earth Science and Disaster Prevention (NIED), and university researchers.

In parallel with the R&D programme, there has also been an effort to make a system for implementing HLW disposal. In the Special Committee on High-Level Radioactive Waste Disposal, AEC, and in the nuclear sub-committee of the Advisory Committee for Energy, Ministry of International Trade and Industry, various aspects of HLW disposal were considered, including social and economic aspects. A law<sup>1</sup> for implementing geological disposal was passed in the Diet in May 2000. Based on the law, an implementing entity<sup>2</sup> for HLW disposal was established in October 2000. The programme then moves from the generic into the site-specific phase. Thereafter we are looking to start operation of the repository between 2030 and the mid-2040s at the latest.

## 2. Status of partitioning and transmutation study

At the same time, recognizing the nature of the radioactive nuclides contained in HLW, the aim since the early days of nuclear energy has been to develop technology either to separate useful elements and nuclides in order to re-use them effectively, or to transmute long-lived nuclides into short-lived or stable – i.e. non-radioactive – forms by irradiation.

In Japan, reference to P&T technology for long-lived and other nuclides first appeared in the *Long-term Programme for Nuclear Research, Development and Utilisation* (or “long-term nuclear programme”) back in 1972. That programme noted the need for research and development in order to ensure effective processing of radioactive waste.

In a 1976 interim report by the AEC’s Technical Advisory Committee on Radioactive Waste, the relationship between P&T technology and the disposal of radioactive waste was specifically pointed out. Specifically, if radioactive nuclides in waste could be appropriately separated into groups, waste management could become more flexible. This is because the amount of radioactive nuclides requiring strict control would be reduced, and treatment and disposal appropriate for the half-life of each group would become possible. In addition, if long-lived nuclides could be transmuted into short-lived ones by nuclear reaction, the burden of long-term waste management could also be reduced.

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<sup>1</sup> Specified Radioactive Waste Final Disposal Act.

<sup>2</sup> Nuclear Waste Management Organisation of Japan (NUMO).

The long-term nuclear programme issued in 1987 stated that P&T technology was very important from the viewpoint of recycling HLW and enhancing disposal efficiency. It also stated that systematic R&D would be carried out jointly by JAERI, the then Power Reactor and Nuclear Fuel Development Corp. (PNC, now JNC) and others.

Under that programme, in 1988, the AEC's Advisory Committee on Radioactive Waste Measures issued a report entitled, *Long-term Programme for Research and Development on Nuclide Partitioning and Transmutation Technology*<sup>3</sup> This can be considered to have been the first systematic R&D programme on P&T technology in Japan. It presented a plan for R&D that ran from 1988 to 2000 and was divided into two phases: Phase I, covering the first four to nine years, which included evaluation of various concepts and R&D on key technologies; and Phase II, covering the next four to nine years, which included engineering experiments on key technologies and demonstrations.

The long-term nuclear programme issued in 1994 stated that each research institute would carry out basic studies on P&T technologies and evaluate each technology at some time in the mid-1990s to determine how to proceed thereafter.

Meanwhile, in 1998, the AEC's Special Committee on the Disposal of High-Level Radioactive Waste released *Basic Concepts in the Disposal of High-level Radioactive Waste*. This stated that, in order to gain public understanding of disposal technology, "research on waste reduction with the aim of achieving safer and more efficient geological disposal, as well as more efficient use of waste, would be carried out on a regular basis". It also said that it was "important to have mechanisms to respond flexibly to any dramatic progress in P&T technology".

Under these circumstances, the AEC's Advisory Committee on Nuclear Fuel Cycle Back-end Policy investigated and considered matters concerning P&T technology for long-lived and other nuclides, based on the evaluation schedule stated in the long-term nuclear programme issued in 1994. The Committee issued a report entitled, *Research and Development of Technologies for Partitioning and Transmutation of Long-lived Nuclide Status and Evaluation Report* in March 2000. The brief summary of this report is as follow.

### **3. Results to date and analyses of current status**

#### **3.1 Elements subject to P&T**

Long-lived (i.e. long half-life). In particular:

- High radiotoxicity due to the emission of rays.
- Fast migration through geological formations via underground water, when disposed of geologically.

Relatively short-lived and heat-generating, producing most of the heat in HLW.

Rare and useful elements.

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<sup>3</sup> Called the "OMEGA Programme" – an acronym for Options Making Extra Gains from Actinides and Fission Products.

## **3.2 Results to date and analyses of current status**

### *3.2.1 The partitioning process*

#### 3.2.1.1 JAERI

JAERI is developing a four-group partitioning process, in which elements in concentrated high-level liquid waste HLLW are separated into four groups: MAs, Tc-platinum group metals, Sr-Cs, and others. Basic experiments using simulated HLLW helped to establish the concept. Test runs with both simulated and actual HLLW on a scale 1/1000 that of an actual plant confirmed the expected capabilities of group partitioning. A recovery rate for MAs of 99.95% or better was achieved.

#### 3.2.1.2 JNC

JNC is developing an improved PUREX process – an advanced version of the conventional reprocessing process – to recover Np. It is also developing a CMPO-TRUEX process to separate MAs from highly concentrated HLLW and an electrolytic extraction method to separate Tc-platinum group and other elements from aqueous reprocessing solutions. In the CMPO-TRUEX process research, it was demonstrated that Am and all nuclides can be recovered to a level of 99.9% or more under standard extraction conditions.

#### 3.2.1.3 CRIEPI

CRIEPI is developing a reductive-extraction process using molten chlorides and liquid metal solvents. Basic data were obtained for the behavior of elements in this type of molten-salt-and-liquid-metal system. Experiments on the separation and recovery of TRUs were carried out using some 10 milligrams of TRUs and some 100 grams of chlorides, which confirmed recovery of more than 99% of the TRUs and adequate separation of TRUs from REs.

### *3.2.2 The transmutation cycle*

JNC and CRIEPI are studying transmutation technology using fast breeder reactors (FBRs). This concept is centered on the use of fast reactors for electricity generation. In this scenario, FBR will take over the role of light-water reactor (LWR) in future, with power generation and the transmutation of MAs and other elements carried out simultaneously by the FBR. In contrast, JAERI's concept is the "double strata fuel cycle", where a dedicated system for transmuting MAs, such as an actinide burner fast reactor (ABR) or an accelerator-driven subcritical system (ADS) is at the centre of the transmutation cycle, allowing a commercial power generation cycle and a transmutation cycle to be developed and optimized independently for their individual purposes.

### *3.2.3 The fuel production process*

#### 3.2.3.1 JAERI

JAERI is developing MA-nitride fuel. The basic data on thermal properties of MA-nitrides necessary to design the fuel, as well as thermal properties of Tc alloys, were obtained. It was confirmed that the MA nitrides could be prepared by means of carbothermic synthesis and uranium nitrides microspheres can be produced via the sol-gel process. In addition, through irradiation tests of U-Pu mixed nitride fuel produced on a trial basis, it was ascertained that the fuel element was intact after a burn-up rate of 5.5% achieved.

### 3.2.3.2 JNC

JNC is developing fuel in which Np and/or Am is added to the MOX fuel that JNC has developed for FBRs. For the addition of Np to MOX fuel, a vibro-packing process is being developed in a joint international research effort. For the addition of Am to MOX fuel, in addition to irradiation experiments being carried out at the experimental fast reactor “Joyo”, remote fuel fabrication facilities, including those to produce pellets and inspect fuel pins, were established and performance tests were carried out.

### 3.2.3.3 CRIEPI

CRIEPI is developing metallic fuel – an MA-content U-Pu-Zr ternary alloy under the international collaborations. Fuel pins have been made on a trial basis and physical characteristics and other basic data have been obtained. It has been determined that an MA content of about 5% does not affect the characteristics of the fuel, and it is expected that MAs can be mixed homogeneously during fabrication of the fuel.

## 3.2.4 *The transmutation process*

### 3.2.4.1 JAERI

JAERI is developing concepts for ADSs and ABRs. Nuclear data on MA nuclides were obtained through international co-operation and were verified while developing a database, carrying out integral experiments and analysing irradiated MA samples. In the development of a proton accelerator for the ADS, major key technologies were developed, and the highest level performance has been demonstrated.

### 3.2.4.2 JNC

JNC is developing a MOX-fuelled FBR. The prototype reactor “Monju” already exists – i.e. construction of a “prototype plant” has been achieved – but the development of other key technologies will be required to add MAs to MOX fuel. Nuclear data on MA nuclides were obtained and evaluated via nuclear reactors and accelerators. Design studies were carried out on acceptable amounts of MAs and rare-earth elements, and on actual fuel loading.

### 3.2.4.3 CRIEPI

CRIEPI is developing the concept of metallic-fuelled FBRs. Regarding nuclear data on MA nuclides, analysis programmes for MA transmutation were developed and analyses were carried out.

## 3.2.5 *Fuel processing*

### 3.2.5.1 JAERI

JAERI is developing a pyrochemical process similar to dry reprocessing. Molten-salt electrolysis of U-nitride, Np-nitride and Pu-nitride on a gram scale were carried out to confirm that transuranic metals can be recovered. For the recovery and recycling of N-15, nitrogen (N<sub>2</sub>) release in molten salts was studied, and it was confirmed that almost 100% of the nitrides can be released in the form of N<sub>2</sub>.

### 3.2.5.2 JNC

JNC is considering the same method as for the partitioning process.

### 3.2.5.3 CRIEPI

CRIEPI is developing molten-salt electrorefining and reductive extraction, which is similar to the partitioning process. Electrorefining forms the main part of the pyro-reprocessing method. Feasibility was confirmed through an in-house study and international joint research, and the process is now at the stage of engineering experiments. Feasibility of the conversion of oxides of spent fuel to metal, and of the spent-salt treatment process, are still to be confirmed.

### 3.2.6 Conclusion

R&D at the three research institutes has resulted in establishment of processes for P&T technology with the expected performance. The aims of Phase I R&D have thus been achieved. R&D in Phase II has experienced some delays, the primary reasons being that Japan is redefining its entire FBR programme, and facilities to handle MAs and other materials have yet to be constructed. In carrying out further R&D, it is important to promote cooperation with domestic and foreign organizations in order that experimental facilities – including those for engineering experiments – can be used efficiently.

## 3.3 Technical issues

The implementation of experiments to demonstrate processes using actual HLLW is an issue common to the three organizations. Also common to JAERI and JNC, which are developing aqueous partitioning processes, are development of a method to more efficiently separate MAs and rare-earth elements, and technologies to reduce the volume of secondary waste. At CRIEPI, where dry partitioning is being developed, the main technical issues in the handling of molten chlorides are material development and molten-salt transport technology. Common issues to the three organisations are preparation of a database on fuel irradiation behavior for performance analysis, and development of fuel fabrication technology.

## 4. Effects and significance of partitioning and transmutation technology

### 4.1 Radioactive inventory in waste

It is a social imperative to minimise, as far as possible, the generation of hazardous waste produced by industrial activities. Reduction of long-term radioactive inventory through the removal of long-lived nuclides from HLW by P&T technology helps to meet this requirement. If, for example, 99% of MAs contained in spent fuel can be removed, the toxicity of the spent fuel after several hundred years following reprocessing will be equal to the toxicity of the same amount of natural uranium as used in the production of the original fuel.

## ***4.2 Effects on geological disposal***

### **4.2.1 Long-term safety**

- Effects on the underground water migration scenario:

Maximum dose can be reduced by about two orders of magnitude by separating and removing 99% of the  $^{135}\text{Cs}$ , and 99% or more of the Np and Am, which are parent nuclides of  $^{229}\text{Th}$ .

- Effects on the human intrusion scenario:

Risks can be reduced by two orders of magnitude by separating and removing 99-99.99% of the actinide elements.

### **4.2.2 Impact on geological disposal site design**

Approximately two-thirds of the heat from HLW is generated by Cs and Sr, and separation and removal of these elements would shorten the required storage period and reduce the size of the site. The HLW storage period could be reduced by separating and removing exothermic nuclides. In addition, disposal site design could be rationalized by, for example, disposing of the HLW in one large cavity rather than in several smaller ones.

## ***4.3 Effective use of resources***

Among the materials in HLW, some can be used effectively as resources. For example platinum group elements are widely used as catalysts to reduce nitrogen oxides in vehicle exhaust gases, and so on. However, prior to actual application, clearance level issues are to be solved.

## ***4.4 Reduction of MA and LLFP inventory, and the times required***

Even if P&T technology is employed, some MAs and LLFPs will remain in the waste, and final disposal of such waste will eventually be necessary. An oxide-fuelled or metallic-fuelled fast reactor can transmute MAs from more than five or six LWRs every year. While an ADS with one quarter of thermal output of a LWR can transmute MAs from more than ten LWRs (about 250 kg per year).

## ***4.5 Generation of secondary waste***

The aqueous partitioning process, like PUREX reprocessing, generates secondary waste. Compared with reprocessing, however, the volume of secondary waste is lower because P&T technology deals with the very limited quantities of HLLW generated by reprocessing.

In the dry-type partitioning process, molten salts and liquid metals are used as solvents, and metallic Li is used as a reducer. These solvents and reducers are less susceptible to degradation by radiation, and may be recycled in the system. But there is almost no experience of using this process on an industrial scale.

To evaluate the secondary waste volume, further investigations and experience using actual materials are needed.

#### ***4.6 Short-term increase in radiation dose***

When P&T technology is employed in the nuclear fuel cycle, exposure dose could increase in the short term as new processes and facilities are introduced and as the volume of MAs and LLFPs to be dealt with in such processes increases. It will be possible, however, to keep exposure dose for workers and the public below the statutory standard, and as low as reasonably possible, by measures such as enhanced shielding.

#### ***4.7 Economy***

R&D to date has focused on basic studies to obtain fundamental data for designing processes and systems, and no one is in a position to make a reliable cost estimate for the P&T technology. Nevertheless, very preliminary cost estimates of three organisations indicate a few percent of LWR electricity generation cost for PT implementation.

### **5. Future research and development**

Modern society demands maximum control of hazardous waste produced by industry, as well as recycling to preserve resources and protect the environment. It goes without saying that the nuclear industry, too, must take all effective measures. P&T technology applied to long-lived nuclides can be useful in, for example, reducing the long-term radioactive inventory in nuclear waste, and it is appropriate that R&D should be carried out on an ongoing basis.

#### ***5.1 P&T technology and the nuclear fuel cycle***

P&T technology is a part of the nuclear fuel cycle. The question of how and in what part of the nuclear fuel cycle P&T technology should be incorporated in order to optimize the cycle, should be considered. The purpose of R&D is to suggest scenarios for introducing P&T systems into the nuclear fuel cycle, and to develop designs and establish key technologies for such systems. Keeping the totality of the fuel cycle in mind, it is necessary to correctly evaluate issues of economy, energy security, and reduction of the radioactive inventory in waste, and to analyse the trade-off among those factors.

#### ***5.2 Research on system design and development of key technologies***

R&D for P&T technology consists of research on system design with the aim of introducing this technology into the nuclear fuel cycle, and development of the key technologies necessary to realise the system. Development of key technologies takes time and should be carried out in a progressive fashion, while at the same time being properly integrated with research on system design.

#### ***5.3 How to proceed R&D in future***

P&T technology based on the use of power-generating fast reactors, and P&T technology based on the double-strata fuel-cycle concept, on which R&D is being carried out in Japan, have their own distinctive features. These two concepts also provide new options for the fuel cycle and it is therefore appropriate at this stage to continue the development of both. The objectives of further R&D are to study scenarios, including a possible blending of these two concepts, in order to introduce a feasible P&T technology system into the nuclear fuel cycle, and to develop the necessary technologies. System

design and P&T introduction scenarios will continue to be studied. According to the scenarios thus defined, and based on the results of R&D to date, small-scale experiments to demonstrate the feasibility of a series of processes will then be conducted. Following this, systems whose feasibility has been successfully demonstrated will be subjected to engineering tests in order to obtain data on their safety. It will be important at this stage to manage the R&D under a system of checks and reviews, and to update the scenarios on a regular basis.

#### ***5.4 Co-operation for R&D***

Although there are differences in the concepts, reactor types and systems, many common issues exist in R&D on P&T technology. JAERI, JNC and CRIEPI should work together in an effort to resolve those common issues, strengthening their co-operation through the sharing of their R&D results. At the same time, it is important to carry out the R&D effectively by, for example, cooperating with other domestic organizations and using their existing facilities. In Western nations, the trend is toward international co-operation in various areas of P&T technology R&D, and the three organisations should actively join such cooperation frameworks and make use of research facilities available overseas. They are also encouraged to exchange information through the existing OECD/NEA framework.

#### ***5.5 R&D schedule and evaluation***

P&T technology is inseparable from the nuclear fuel cycle, and it is therefore appropriate to conduct R&D in this area on a time schedule compatible with nuclear fuel cycle R&D. At present, feasibility study on commercialised FBRs and related fuel cycle system is being carried out under the collaborative efforts of JNC, electric utilities, CRIEPI and JAERI. In this study, R&D scenarios toward commercialization of fast reactor system will be reviewed by about 2005. Thus, around the year 2005 is deemed to be an appropriate time to reconsider all R&D scenarios of PT including the use of FBRs for transmutation together with power generation, and the double-strata fuel cycle. Thereafter, progress, results and R&D policy will be checked and reviewed every five years or so. Evaluations of P&T technology system concepts, and reviews of introduction scenarios, should also be conducted.

### **6. Concluding remarks**

P&T technology belongs to a world quite different from that of ordinary chemical reactions in the sense that, in P&T, materials are transformed at the atomic level. Its potential is not limited simply to transmutation. In addition, its development raises issues that will be difficult to resolve with existing technology alone, overcoming these difficulties could lead to other exciting technological breakthroughs. This, indeed, should be one reason for young engineers scientists to want to become involved in the field. Research on this kind of advanced technology can be expected to make a great contribution to revitalizing nuclear research generally. P&T technology research should be actively promoted in order to nurture the development of human resources in the nuclear field.

In doing this, however, it is important to create an environment in which innovative ideas can be adopted without “interference” from existing systems. P&T technology requires open-minded R&D.

Inspired by Japan’s OMEGA programme, many similar programmes have been established around the world – in France, other European countries, and the United States. It is expected for Japan to play an important on-going role in this area – and to do so as part of her international contribution, while conducting timely evaluation of R&D progress.

**R&D scheme for partitioning and transmutation of long-lived radioactive waste in Japan**

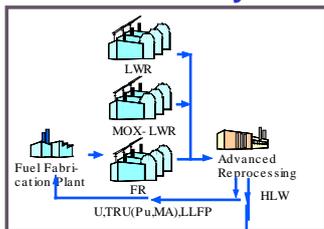
**JNC, CRIEPI**

- † **Advanced fuel cycle**
- † Commercial FR for transmutation
- † Oxide fuel, Metal fuel
- † 2~5% MA content (homogeneous)

**JAERI**

- **Double-strata fuel cycle concept**
- Combination of a power reactor fuel cycle & an independent P-T cycle
- † Dedicated systems for P-T
- † Accelerator-Driven System (ADS)

**Advanced fuel cycle**



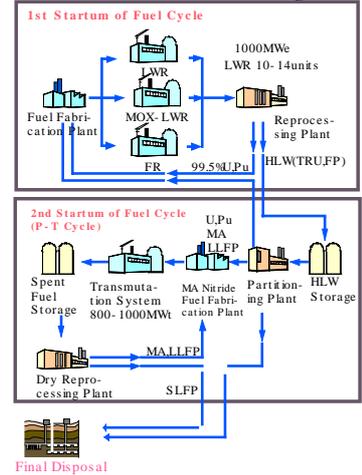
MA ; Minor Actinide  
LLFP; Long-lived FP  
SLFP; Short-lived FP

Common technologies

**R&D items to be developed in collaboration**

- † Separation chemistry
- † Reactor physics
- † Fuel basic property
- † Irradiation test
- † Nuclear data

**Double strata fuel cycle**



Final Disposal

Annex 2

		JAERI	JNC	CRIEPI
Elements subject to P&T	Partitioning	MAAs (Np, Am, Cm), Pu, Tc Platinum group (Ru, Rh, Pd), Sr, Cs	MAAs (Np, Am, Cm), Pu, Tc Platinum group (Ru, Rh, Pd)	MAAs (Np, Am, Cm), Pu
	Transmutation	MAAs (Np, Am, Cm), Tc, I	MAAs (Np, Am, Cm), Pu, Tc, I	MAAs (Np, Am, Cm), Pu
Partitioning process		4-group partitioning process (wet process)	Advanced reprocessing nuclide partitioning system (wet process)	Dry process
	MA separation	DIDPA extraction	CMPO/TRUEX process Improved PUREX process	Reductive extraction (molten salt/liquid metal)
	Tc-Platinum group	Precipitation by de-nitration	Electrolytic extraction	–
	Sr-Cs	Column absorption with inorganic ion exchangers	–	–
Transmutation cycle		Double strata fuel cycle	MOX fuelled FBR	Metallic-fuelled FBR
	Fuel type	MA-nitride fuel	MA-MOX fuel	U-Pu-Zr ternary alloy
	Transmutation process	Accelerator driven sub-critical system (ADS) Actinide burner fast reactor(ABR)	FBR	FBR
	Fuel processing	Molten-salt electrolysis (pyroprocess)	Wet process	Molten-salt electro-refining and reductive extraction

*Annex 3*

**Members of the Advisory Committee on Nuclear Fuel Cycle Back-end Policy  
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Yumi Akimoto	President, Mitsubishi Materials Corporation
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