# ACTIVITIES ON R&D OF PARTITIONING AND TRANSMUTATION IN JAPAN

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## Abstract

In Japan, since plutonium is to be used as a nuclear fuel material under the nuclear energy policy, the objective of partitioning and transmutation (P&T) is oriented to removal and transmutation of mainly MA and long-lived fission products. Under the OMEGA (option making of extra gain from actinides and fission products transmutation) programme, on the basis of the double-strata fuel cycle concept JAERI (Japan Atomic Energy Research Institute) continues to carry out the development of partitioning processes, nitride fuel technology, and a basic study to support the ADS (Accelerator-driven system) development. JNC (Japan Nuclear Cycle Development Institute) bears the research and development of P&T technology of in the course of fast reactor cycle. In the present paper, these activities on the research and development of P&T are introduced.

## Introduction

In Japan, since plutonium is to be used as a nuclear fuel material under the nuclear energy policy, the objective of P&T is oriented to removal and transmutation of mainly MA and long-lived fission products.

The progress of the OMEGA programme of JAERI, JNC and CRIEPI were reviewed in 1999 by the Atomic Energy Commission's Advisory Committee on Nuclear Fuel Cycle Back-End Policy and the report was issued in March 2000. Necessity of such R&D works was concluded as future system design and the development of the implementation scenario of P&T, basic experiments to demonstrate the feasibility of the processes, and engineering scale experiments in order to obtain safety data of these systems.

The R&D programme has been jointly stimulated by the collaborative efforts of JAERI, JNC and CRIEPI.

The R&D areas covered by the OMEGA Programme are as follows:

- Physical and chemical properties of MA and fission products.
- Partitioning of radioactive elements from HLW of reprocessing process.
- Transmutation: nuclear and fuel property data of MA, system design studies, reactor fuel and accelerator target development, development of high power accelerator for transmutation.

Under the OMEGA programme, JAERI continues to carry out the development of partitioning processes, nitride fuel technology, and a basic study to support the ADS development. The answer to this lies in the following facts: [1] ADS has flexibility for Nuclear Waste Transmutation in some scenarios of nuclear reactors considered in future: For instance, In case of RMWR and /or FBR introduction such as $UO_2$-LWR / MOX-LWR / RMWR / FBR, ADSs can transmute MAs from LWRs, and (Am,Cm) from RMWR / FBR and co-exists with RMWR/FBR. [2] ADS is the dedicated MA transmutation system and it is independent from Commercial Fuel Cycle. P&T with ADS reduce burden for commercial reactors in economy and safety. MAs are confined in one small P&T cycle site and not extended in commercial fuel cycle. [3] ADS consists of sub-critical system with external neutron sources using high-intensity proton accelerator, and one has control and flexibility in the design and operation to transmute large amounts of MAs in contract to critical reactors. As a result, ADS provides high efficient and safety transmutation system. The R&D for P&T technology in JAERI have been carrying out on the basis of the double-strata fuel cycle concept. The JAERI`s activities are summarised on the recent technical achievements for analyses of mass flow and cost estimate for a double-strata fuel cycle, lead-bismuth technology, experimental facilities for ADS technology demonstration, partitioning and fuel processes.

In recent years, new attempts to seek for the better concepts of the nuclear fuel cycle have been made in Japan. Under these circumstances, JNC, whose basic policy is to conduct partitioning and transmutation of MA and FP in the course of fast reactor cycle, has tried to reconstruct the basic planning of partitioning and transmutation in view points of the development goal, the nuclides to be partitioned and to be transmuted, and the development schedule. As for the goal of partitioning and transmutation technology, step by step approach that consists of three steps, will be adopted. Nuclides to be partitioned and to be transmuted are selected in accordance with each goal in the following three view points; radio activity/radiotoxicity, improvement of repository, and effective use. The selected nuclides for the first step, for example, are U, Np, Pu, Am, Cm, Tc, Sr, Cs, Mo, I, and Pd, with the recovery rate of 99% for U, Np, Cm, Tc, and 99.9% for Pu, Am. The major subjects of partitioning and transmutation in order to accomplish the goal are listed up with the development schedule.

**Activities for R&D of P&T based on ADS**

Under the OMEGA programme, JAERI continues to carry out the development of partitioning processes, nitride fuel technology, and a basic study to support the ADS development. R&D activities on ADS, recently more and more university and industry groups have got involved in this area, not only from nuclear energy community but also nuclear physics and accelerator communities.

The R&D for Partitioning and Transmutation (P&T) technology in JAERI have been carrying out on the basis of the double-strata fuel cycle concept [1,2] as shown in Figure 1. The JAERI`s activities are summarised on the recent technical achievements for the nuclear transmutation, partitioning and fuel processes. The main R&D items for the ADS are shown in Figure 2.

***Analysis of mass flow and cost for double-strata fuel cycle***

As the commercial reactor fuel cycle is assumed to be sustainable in a significantly long term, the following different reactors and fuel cycles are considered: Co-existing scheme of ADS in Pu-utilisation fuel cycles with MOX-LWRs and FBRs. Furthermore, impact assessment of P&T cycle on geological disposal is studied, and the results are shown in Figure 3. The performance of the ADS considered here [3] is 800 MWt and can transmute MA and $^{129}$I produced per year from about 10 PWRs of 3 400 MWt.

Figure 1. **Concept of the double strata fuel cycle based on ADS**

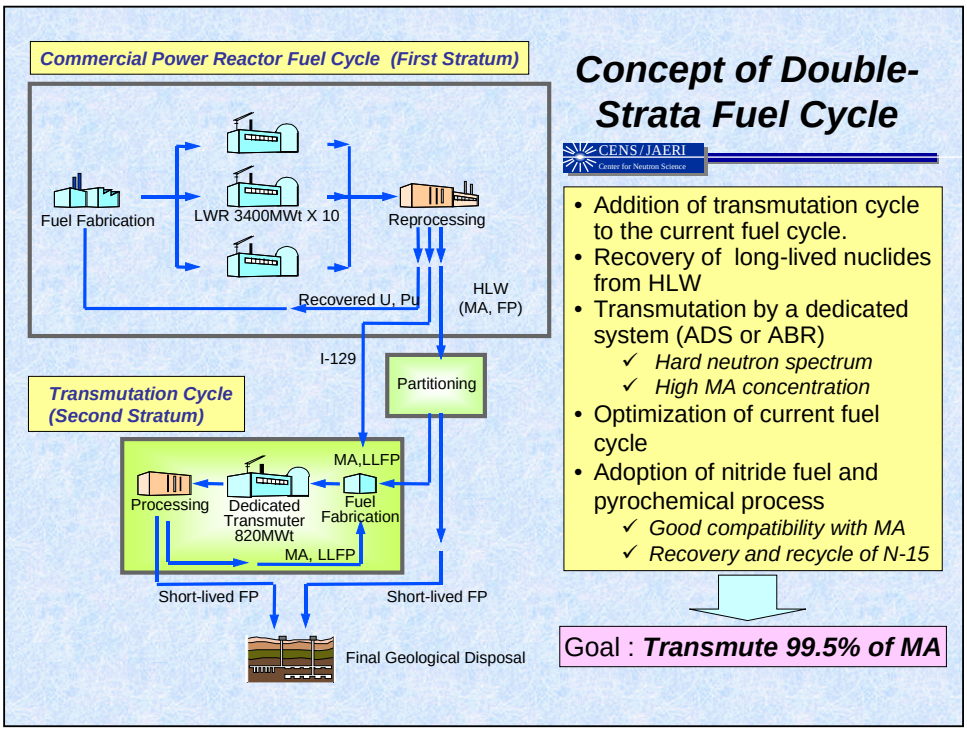


Figure 2. **Main R&D items for ADS**

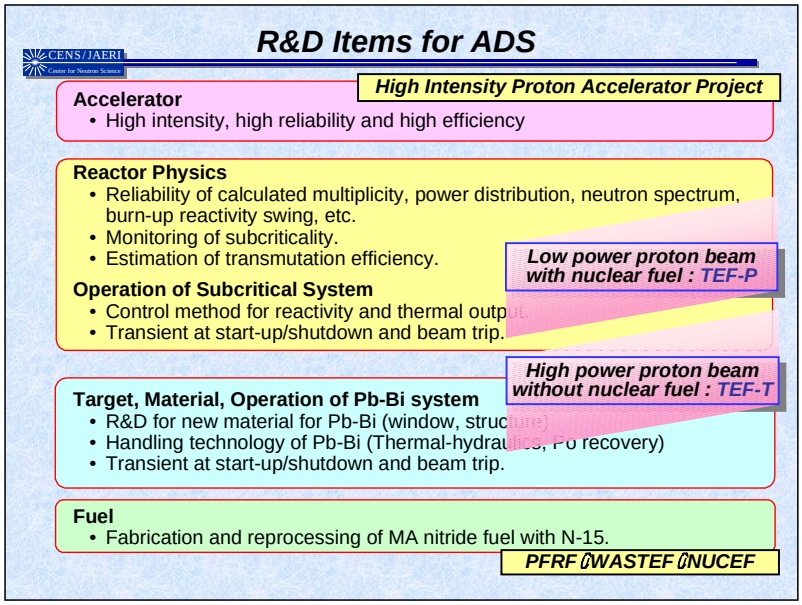
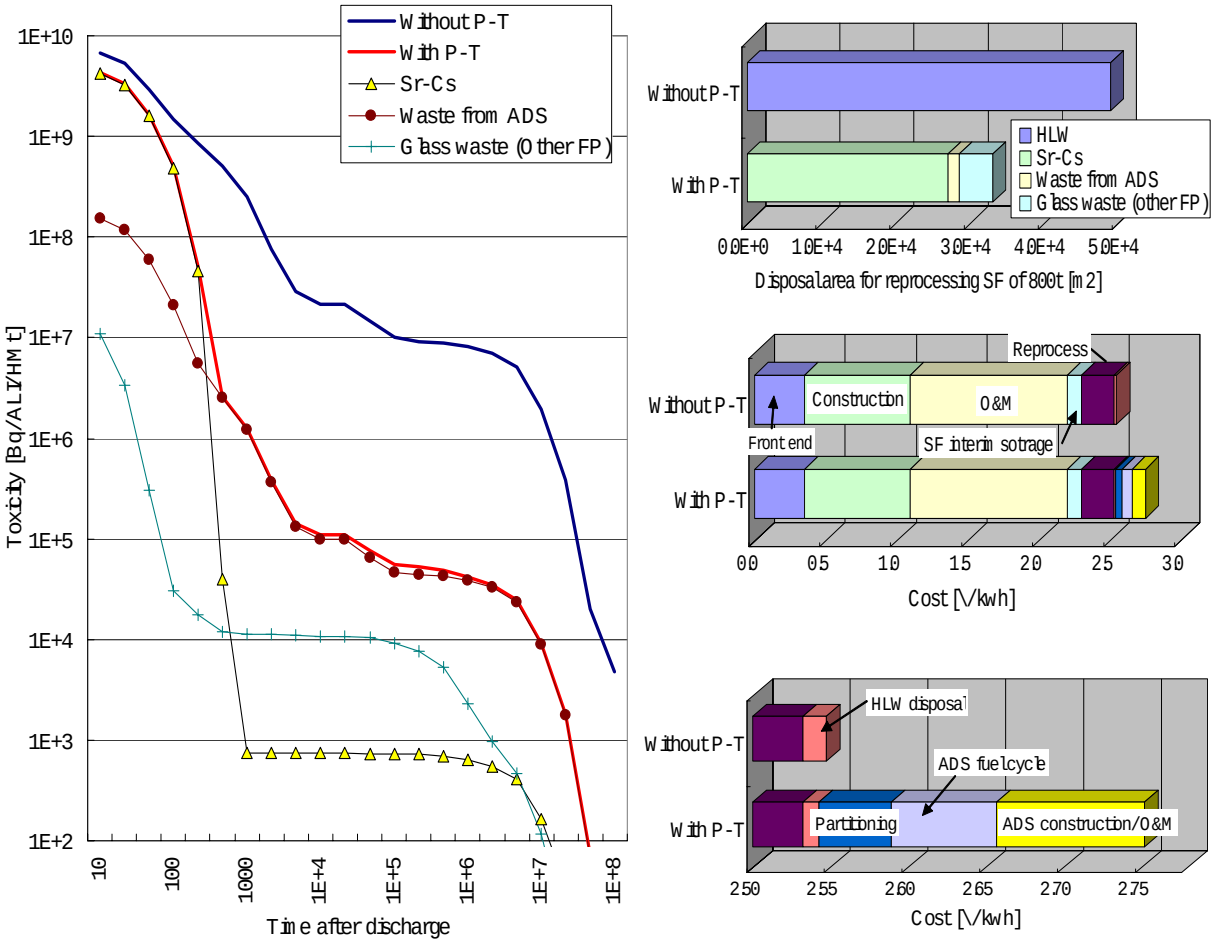



Figure 3. **Radiotoxicity reduction, impact on geological disposal and cost estimate by double-strata cycle scheme with ADS introduction for P&T**



### *Lead-bismuth technology and material development*

For target, materials and operation of Pb-Bi system, a material test loop and stagnant test devices are installed for Pb-Bi technology development and they are in operation. The statistic corrosion tests were performed after 3 000 h at 450°C. The loop corrosion test (Figure 4) was conducted under the temperature of 450°C, EMP power of 5 L/min. and velocity of 1m/s. [4] For irradiation damage, PIE is underway for samples irradiated in TIARA at JAERI/Takasaki, where triple beams combining protons with helium and iron ions were used to simulate heavy irradiation. PIE for irradiation at SINQ is also in progress

### *Experimental facilities for ADS technology demonstration*

The experimental programme for development and demonstration of accelerator-driven transmutation technology has been carried out under the project plan of the High Intensity Proton Accelerator and the OMEGA Programme at JAERI. [5,6] Pre-conceptual design study is being made for a transmutation experimental system as shown in Figure 5. [7] The major areas of technology to be tested and demonstrated are sub-critical reactor physics, system operation and control, transmutation, thermal-hydraulics, and material irradiation.

Figure 4. **Lead-bismuth test loop at JAERI**

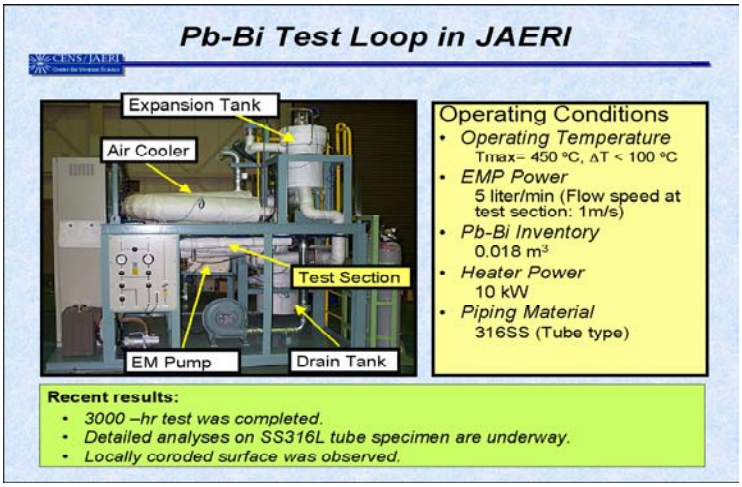
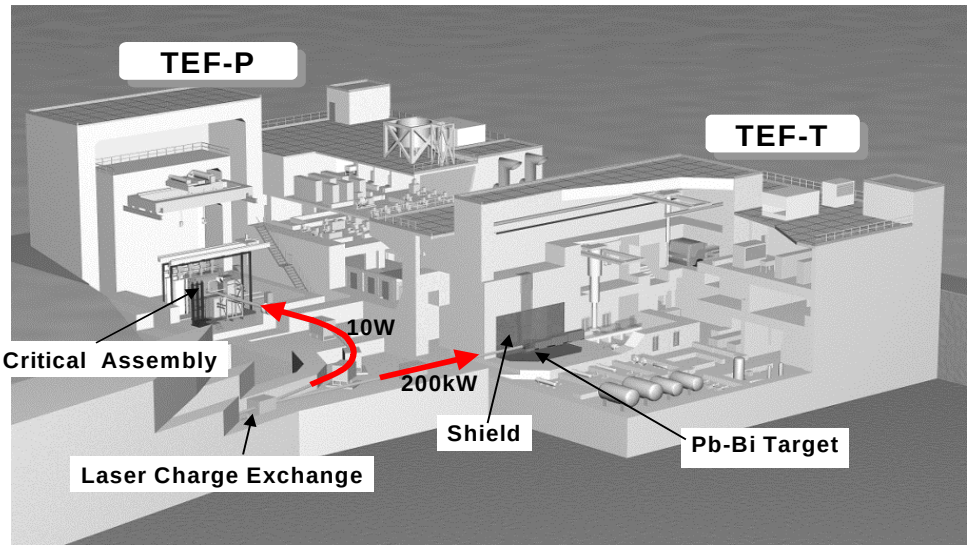


Figure 5. **Transmutation experimental facilities with sub-critical experiment and Pb-Bi target experiment**




***Partitioning technology***

The developed 4-Group Partitioning Process with DIDPA was tested with concentrated real HLW. Objective elements, Am, Cm were separated more than 99.98%, and Np more than 99.95%. As a modification effort of the present process, a more powerful ligand, tridentate diglycolamide (DGA), has been studied to extract actinides. From fundamental studies, TODGA [8] was selected as the most proper DGA extractant. Presently, a new extractant, TODGA, is being studied for the modification of the TRU separation step in order to make the process more effective and more economical.

***Nitride fuel and pyroprocessing***

JAERI's ADS will use (Pu, MA)N fuel diluted by inert matrices such as ZrN and TiN at the initial loading. Then, (Pu,Zr)N and PuN+TiN pellets have been fabricated and fuel pins containing the pellets are to be irradiated at JMTR as shown in Figure 6. Electrorefining of UN, NpN and PuN has been demonstrated up to now. On the other hand, that for AmN is planned from 2003 at TRUHITEC of JAERI Tokai. [9]

***Road map for ADS development***

P&T scenario with ADS transmutor is innovative and flexible system (coexisting with Pu fueled reactors such as MOX-LWR, RBWR and FBR) for reducing high-level wastes. After the basic R&D mentioned above, an experimental ADS with 80MW thermal power is considered to be planed in late 2010s to demonstrate the engineering feasibility of the ADS as shown in Figure 7. The experimental ADS will be operated by MOX fuel at first and gradually altered to MA nitride fuel.

Figure 6. **Nitride fuel pellets fabricated for pin irradiation at JMTR**

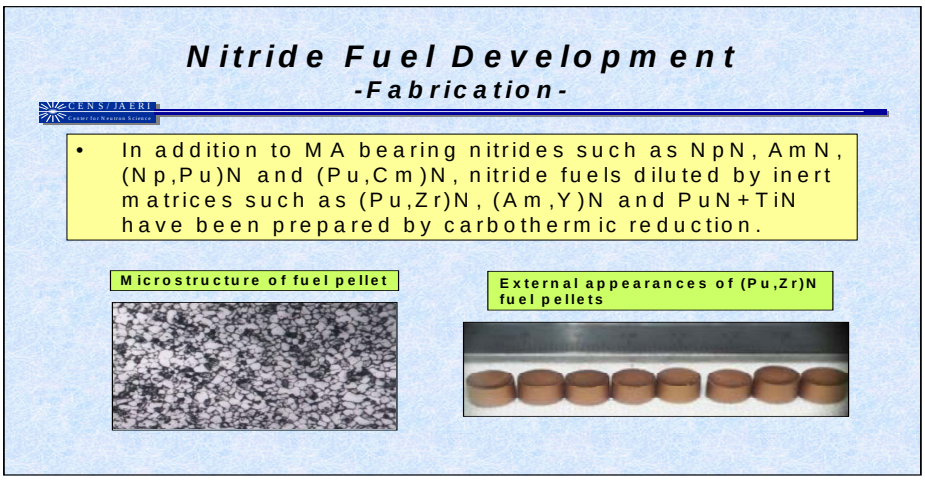
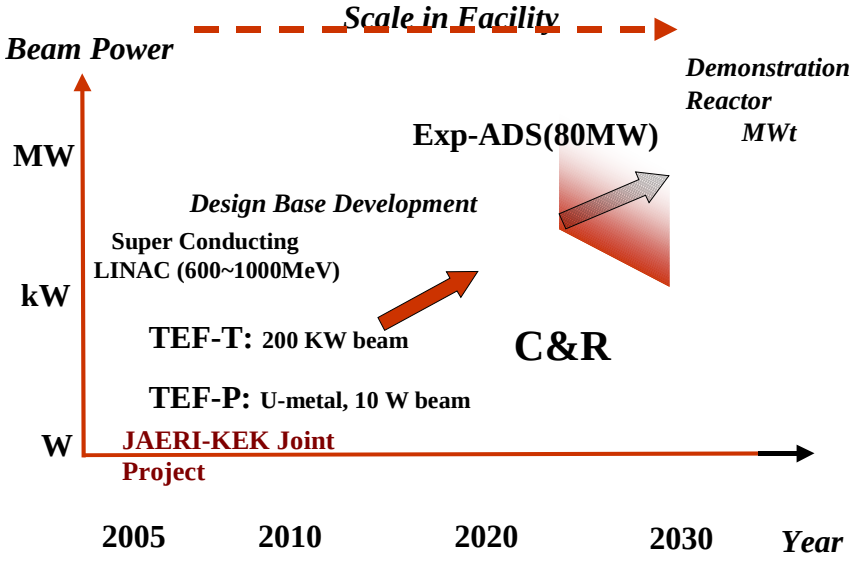



Figure 7. **Road map for ADS development at JAERI**




**Activities for R&D of P&T based on FBR**

***JNC's basic planning of P&T***

JNC, whose basic policy is to conduct partitioning and transmutation of MA and FP in the course of fast reactor cycle, [10] has tried to reconstruct the basic planning of the partitioning and transmutation studies and has tried to clarify the development goals and to clarify the nuclides to be partitioned and to be transmuted. The basic standpoints for the P&T are as following.

MA will be transmuted, but at the same time, will be recycled as a part of resources in the same manner as Pu. 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. Target fuel assembly of MA will not be studied, since there exists no necessity of heterogeneous loading of MA, from following reasons, in the case of fast reactor cycle at present.

- There exists no major problem both in core and fuel characteristics if the MA content in the core fuel of homogeneous loading is less than 5%, [11] while fast reactor, of which equilibrium MA content is around 1%, can accept and burn MA recovered from LWRs satisfactory with the MA content less than 5%.
- Target fuel assembly of MA has a lot of uncertainties to be ensured in recycling including fuel fabrication. [12]
- MA addition to the core fuel will contribute to non-proliferation.

FPs will be classified into four categories, such as the transmutation in the form of target assembly after separation ($^{99}$Tc, $^{129}$I, $^{135}$Cs, etc.), the cooling storage after separation ($^{90}$Sr, $^{137}$Cs, etc), the waste as stable elements after separation (Mo, Nd, Ba, etc.), and. the effective utilisation after separation (Ru, Rh, Pd, etc.). Studies will be carried out based on element separation, though studies for isotope separation will be started after feasibility of isotope separation is confirmed.

***Development goal***

The significance of P&T is to decrease the radioactive waste itself, and in this sense P&T plays an important role together with repository technology complementary fulfilling each other. Furthermore, P&T have possibility to contribute improving the acceptability of radioactive waste through the effective utilisation of FPs represented by rare useful elements.

A step by step approach, consists of three steps, to reach the goal of partitioning and transmutation technology will be adopted as shown in Table 1. The first step is supposed to be able to attain by the present technology and on the extension of it. The second step is such a goal that is expected to be able to realise the engineering feasibility through the progress of science technology in future, although the engineering feasibility is not sufficiently foreseen at present. It will need revolutionary technology or breakthrough, since it will be difficult on the extension of the present technology, in order to attain the second step goal. The ultimate is the ideal and shows the direction to proceed.

Table 1. **Development goal of P&T technology**

| Step | Goal |
|---|---|
| First Step | The radioactivity and the radiotoxicity released from the system should be decreased to 1/100 of those of LWR one-through at the time of 1 000 years later. Furthermore, the partitioning and transmutation of FPs which contribute to reduce the repository burden (1/2) and to reduce the dose risks (1/10), and the partitioning of rare elements which can be effectively utilised should be executed. |
| Second Step | The radioactivity and the radiotoxicity released from the system should be decreased to 1/1 000 of those of LWR one-through after 100 years. Furthermore, the partitioning and transmutation of FPs which contribute to reduce the repository burden (1/5) and to reduce the dose risks (1/100), and the partitioning of rare elements which can be effectively utilised should be executed. |
| Ultimate | The radioactivity and the radiotoxicity released from the system should be decreased to those of natural uranium within one generation so as not to leave the burden of the radioactive waste to the future generation.<br>Furthermore, the partitioning and transmutation of FPs which contribute to reduce the repository burden (1/10) and to reduce the dose risks (1/1 000), and the partitioning of rare elements which has possibility of improving the acceptability of radioactive waste through the effective utilisation should be executed. |

Table 2. **Nuclides to be partitioned and to be transmuted**

| Step | Radioactivity and radiotoxicity view point | | Repository view point | | Effective utilisation view point | |
|---|---|---|---|---|---|---|
| | Radioactivity | Radiotoxicity | Repository capacity | Dose risk | Chemical use (Catalyst etc.) | Radio-chemical use |
| First step | U,Np,Cm (99%)*<br>Pu,Am (99.9%)<br>Tc99 (99%) | U,Np,Am,Cm (99%)<br>Pu (99.9%) | (decrease to 1/2)<br>Sr, Cs (90%)<br>Mo (80%) | (decrease to 1/10)<br>I (99%) | Pd , Tc | Heat source: Cs,Sr<br>Radiation source: Cs |
| Second step | U,Np,Am,Cm (99.9%)<br>Pu (99.99%)<br>Se79,Pd107, Sn126 (99%)<br>Cs135,Zr93,Tc99 Cs137,Sr90, Sm151 (99.9%) | U,Np,Cm (99.9%)<br>Pu,Am (99.99%)<br>Cs137,Sr90 (99%) | (decrease to 1/5)<br>Sr, Cs, (99%)<br>Mo, Nd, Ru, Rh, Ce, Pr, Ba, La, Te, Gd, Rb, Y, Pd | (decrease to 1/100)<br>I (99.9%)<br>C14, Cl36 (1/10) | Pd, Tc, Ru, Rh, Se, Te | Cs, Sr, Ru, Rh, Pd |

recovery rate in the system

| | |
|---|---|
| Ultimate | The radioactivity and the radiotoxicity released from the system should be decreased to those of natural uranium within one generation so as not to leave the radioactive waste burden to the future generation.<br>Furthermore, the partitioning and transmutation of FPs which contribute to reduce the repository burden (1/10) and to reduce the dose risks (1/1000), and the partitioning of rare elements which has possibility of improving the acceptability of radioactive waste through the effective utilisation should be executed. |

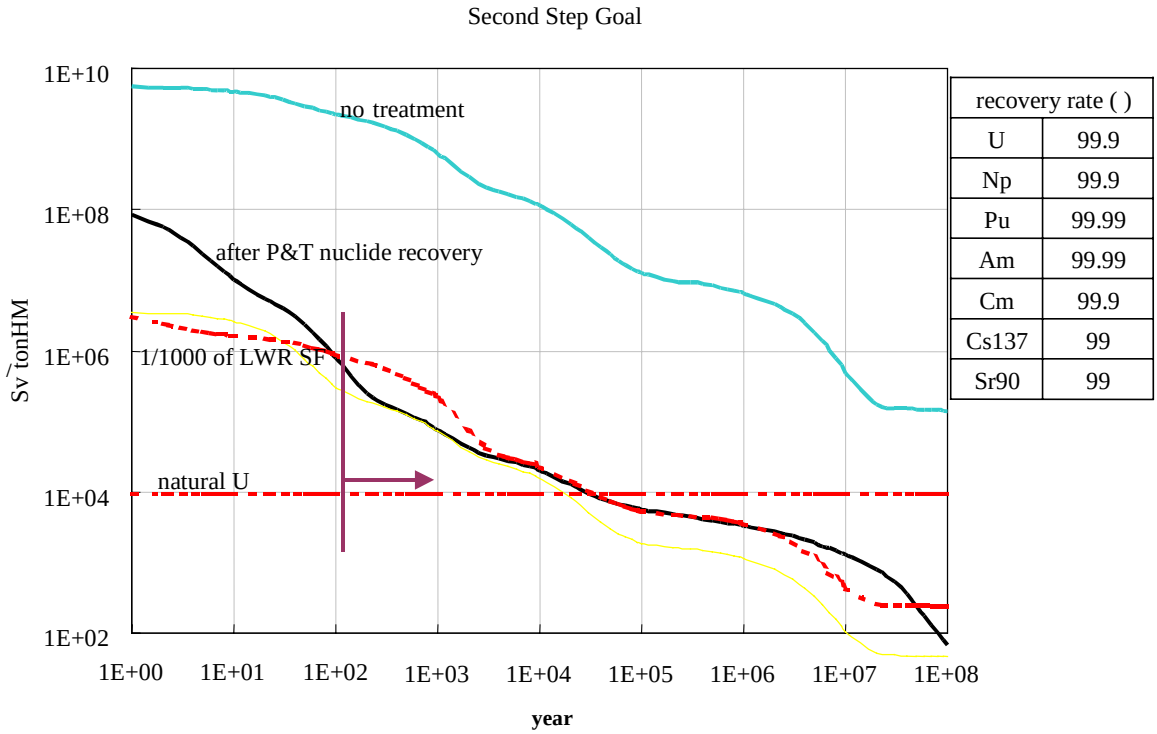
***Nuclides to be partitioned and to be transmuted***

Nuclides to be partitioned and to be transmuted have been selected in following three view points, “radioactivity and radiotoxicity view point”, “repository view point” and “effective utilisation view point”, corresponding to the each step of the development goal as shown in Table 2. Values in parentheses mean the expected whole recovery rate in the system.

The main nuclides to be partitioned and to be transmuted are TRU in the ratiotoxicity view point, though LLFPs are added in the radioactivity view point. The recovery rate of 99.9% for Plutonium and Americium and 99% for other TRU are necessary in the first step, while one order higher recovery rates are required in the second step.

The relationship between the radiotoxicity after P&T nuclides recovery and the aimed radiotoxicity is presented in Figure 8 as one example of selection basis for the second step goal. The curve indicated as “no treatment” presents the radiotoxicity of spent fuel from fast reactor cycle, while the curve indicated as “after P&T nuclide recovery” means the radiotoxicity of the nuclides which leak to outside the system after nuclides, listed in a table on the right hand side of Figure 8, were separated and recovered with the recovery rate shown in the table. The curve indicated as “1/1 000 of LWR SF” corresponds to the development goal of the second step, while the curve “natural U” corresponds to the ultimate development goal. The curve “after P&T nuclide recovery” should be below the curve “1/1 000 of LWR SF” after 100 years in order to satisfy the second step goal, but not necessary be below the curve “1/1 000 of LWR SF” after it cleared the natural U level. In Figure 8, the curve “after P&T nuclide recovery” satisfies the second step goal, so that the nuclides listed in a table on the right hand side have been selected as nuclides to be partitioned and to be transmuted in the second step.

Figure 8. **The radiotoxicity after P&T nuclides recovery and the aimed radiotoxicity**



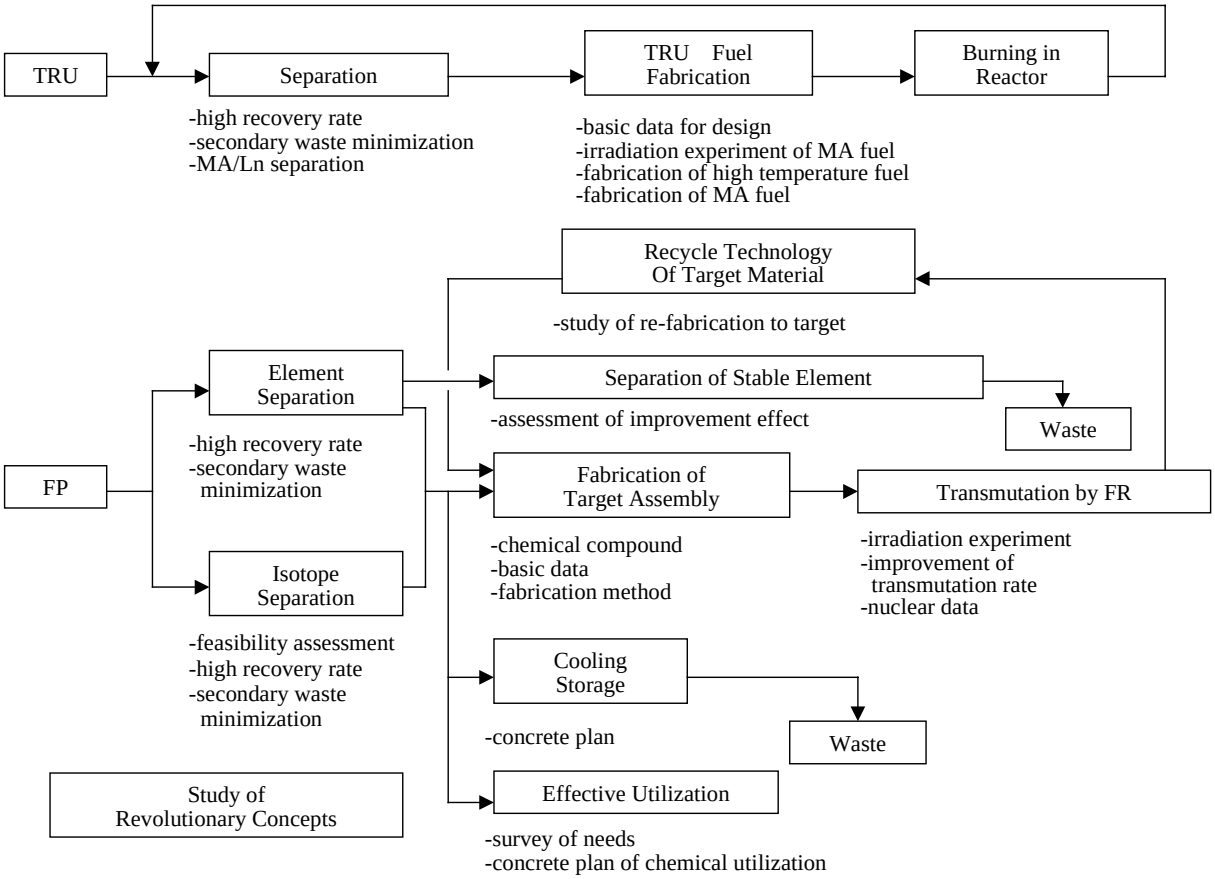

In the repository view point, Strontium and Cesium, which are heat generation nuclides, and Molybdenum, which has bad influence on the durability of vitrified HLW, have been selected in order to increase the repository capacity, while Iodine has been selected to decrease the dose risks, since Iodine has the biggest influence on the dose risks due to its high solubility and low adhesion characteristics to a geologic layer. [13] As for the second step, major stable elements, which occupy about 60% of whole fission products and may not be necessary to be included in vitrified HLW, are selected. Furthermore, Carbon-14 and Chlorine-36 are selected, since they become dominant in dose risks after the one decade reduction of Iodine. Concerning the selection basis, further discussion will be expected in future adopting new knowledge.

Such nuclides have been selected in the effective utilisation view point that Palladium, Technetium, Ruthenium, etc. in expectation for chemical use as catalyst etc., and Strontium, Cesium, etc. for utilisation as heat source and radiation source.

FP nuclides listed in the first step are supposed to be based on element separation, while part of FP nuclides listed in the second step may need isotope separation.

Research and development plan to partition and to transmute the selected nuclides has been made. Major subjects of P&T technology, together with the outline of P&T flow, can be summarised as shown in Figure 9. Residual subjects have been listed up after the present status for the major subjects in Figure 9 has been examined. The research and development plan has been produced considering the listed up residual subjects for the each nuclide in the first step. The each nuclide means TRU, Sr, Cs, I, Tc, Mo and Pd. For an instance, the development schedule of P&T technology for Iodine is presented in Figure 10. Target fabrication of Iodine, recycling technology of Iodine, and verification of analysis method will be established within ten years.

Figure 9. **Major subjects of P&T technology**

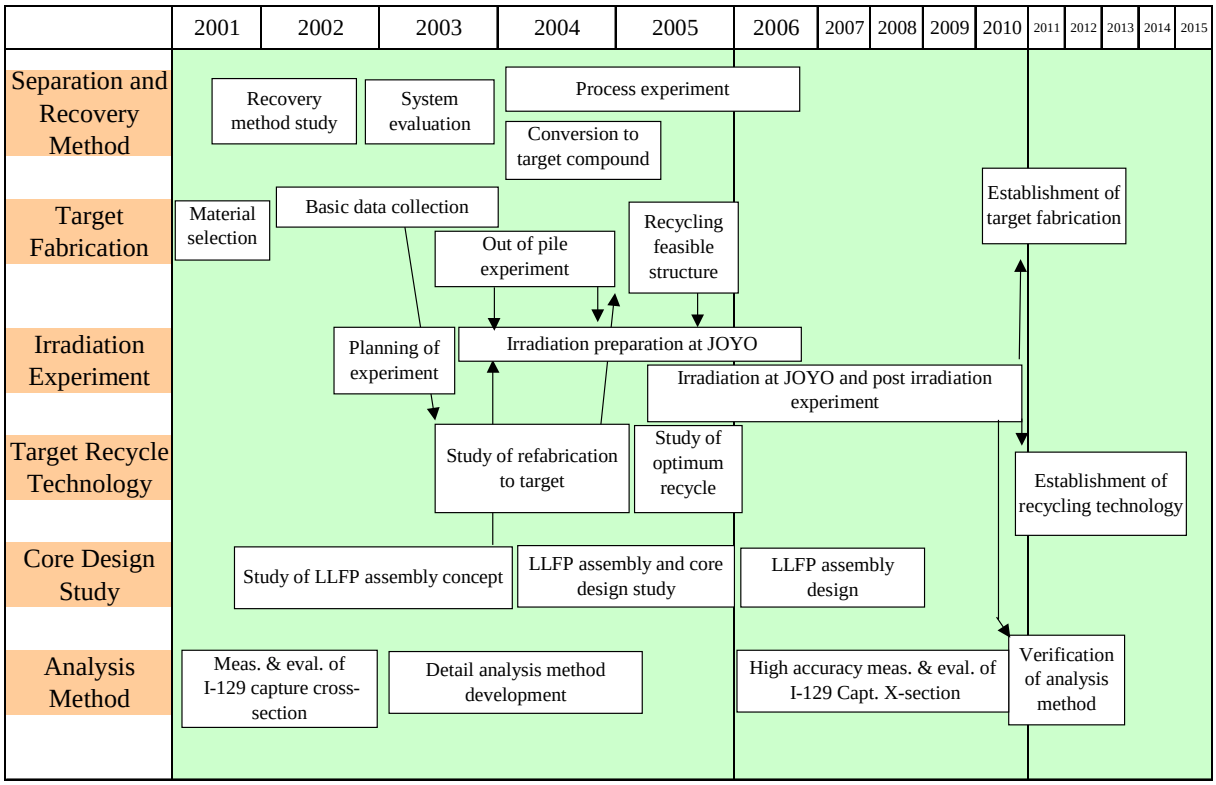



***Conclusions***

P&T scenario with ADS transmutor is innovative and flexible system (coexisting with Pu fueled reactors such as MOX-LWR, RBWR and FBR) for reducing high-level wastes. After the basic R&D mentioned above, an experimental ADS with 80MW thermal power is considered to be planed in late 2010s to demonstrate the engineering feasibility of the ADS. The experimental ADS will be operated by MOX fuel at first and gradually altered to MA nitride fuel.

In P&T scenario with FBR, JNC has showed the step by step approach, consists of three steps, to reach the development goal of partitioning and transmutation technology, and has clarified the nuclides to be partitioned and to be transmuted in following three view points, “radioactivity and radiotoxicity view point”, “repository view point” and “effective utilisation view point”, corresponding to the each step of the development goal. Furthermore, research and development plan to partition and to transmute the selected nuclides has been made.

Figure 10. **Development schedule of P&T technology for iodine**

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