# RECENT TOPICS IN R&D FOR THE OMEGA PROGRAMME IN JAERI

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## Abstract

The R&D for partitioning and transmutation (P&T) technology has been carried out in Japan since 1988 under the OMEGA (Options for Making Extra Gains from Actinides and fission products) programme. In this programme JAERI has proposed the double-strata fuel cycle concept as a partitioning and transmutation system for long lived radioactive nuclides. The system consists of three technical areas or processes of the partitioning, nuclear transmutation and fuel processes. This paper summarises the JAERI's activities on these topics, focusing on the recent technical achievements in each process.

## 1. Introduction

The double-strata fuel cycle concept has been proposed by JAERI as a partitioning and transmutation system for long lived radioactive nuclides. Mukaiyama *et al.*, reviewed the activities in JAERI for research and development of this concept [1]. The system consists of the following three technical areas or processes, the partitioning, nuclear transmutation and fuel processes.

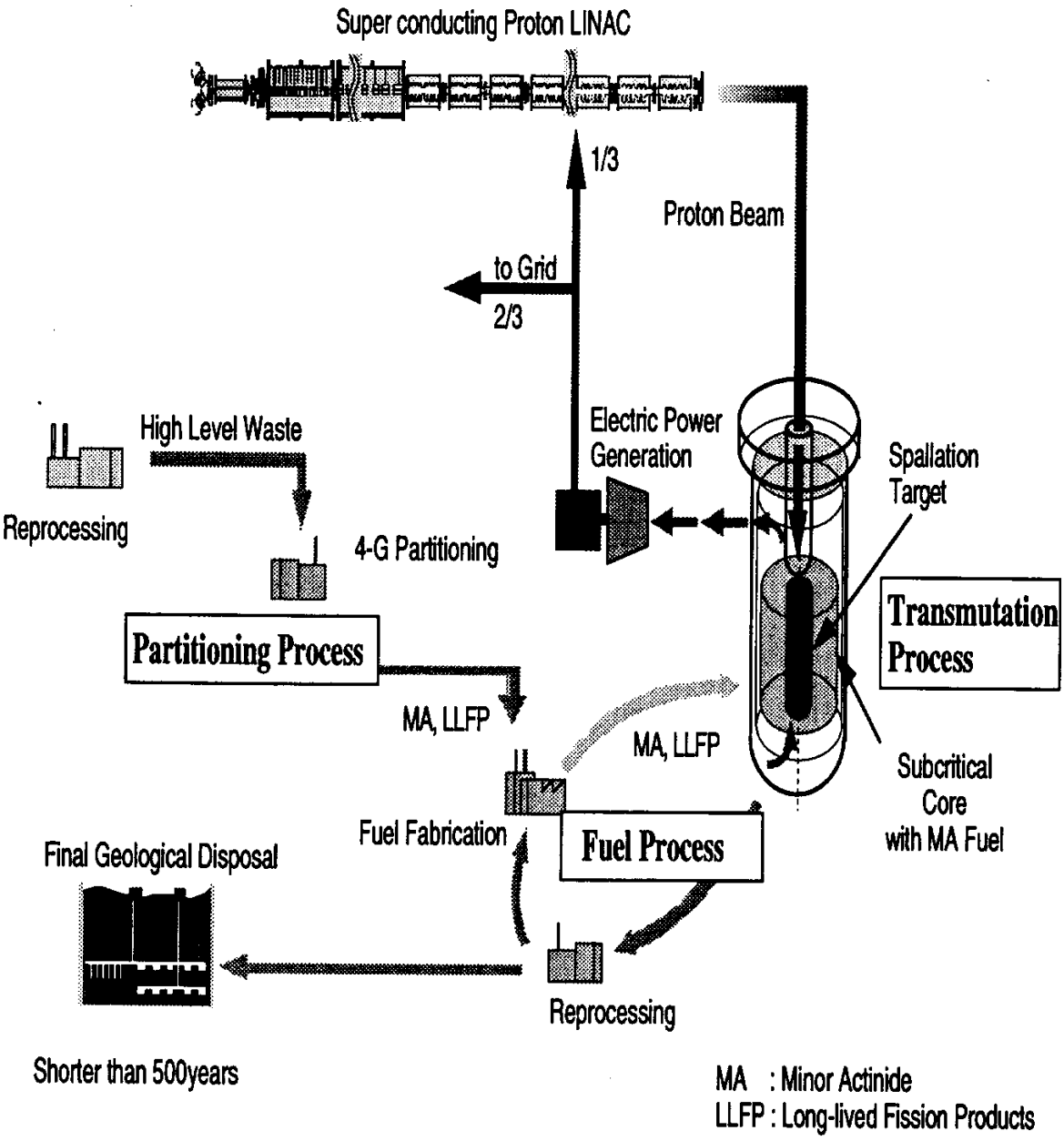
- ***Partitioning process*:** The four-group partitioning process (4-GPP) has been developed to separate the elements in high-level liquid wastes (HLLW) into transuranium elements (TRU), Tc and Platinum group metals (PGM), Sr-Cs group, and others. TRU are separated by extraction with diisodecylphosphoric acid (DIDPA), Tc and PGM by precipitation through denitration, Sr and Cs by adsorption with inorganic ion exchangers (titanic acid and zeolite). A hot verification test was performed using concentrated real HLLW. As a modification effort of the present 4-GPP, a more powerful ligand, tridentate diglycolamide (DGA), has been studied to extract actinides directly from HLLW. From fundamental studies, tetraoctyl 3-oxapentandiamide (TODGA) was selected as the most proper DGA extractant.

- ***Transmutation process*:** A lead-bismuth cooled accelerator driven system (ADS) with nitride fuel has been proposed as a dedicated transmutation system to be deployed into the second stratum. The 800 MWt plant has a pool-type configuration and a power conversion system operated on a saturated steam cycle.

- ***Fuel process*:** Nitride is suitable for the fuel material for MA transmutation from the viewpoint of supporting hard neutron spectrum and heat conduction ability. In addition, actinide mononitride with NaCl-type structure will have a mutual solubility leading to the flexibility of fuel composition. Pyrochemical processing has several advantages over wet process in case of treating MAs concentratedly with large decay heat and fast neutron emission. One of the drawbacks of nitride fuel is that nitride with $^{15}N$ enriched nitrogen must be used to minimise the $^{14}C$ production. But the pyrochemical process has the practical feasibility of recovering expensive $^{15}N$.

In this paper, the recent JAERI's activities in the P&T technology development are reviewed by focusing on the major technical achievements in each process shown in Figure 1.

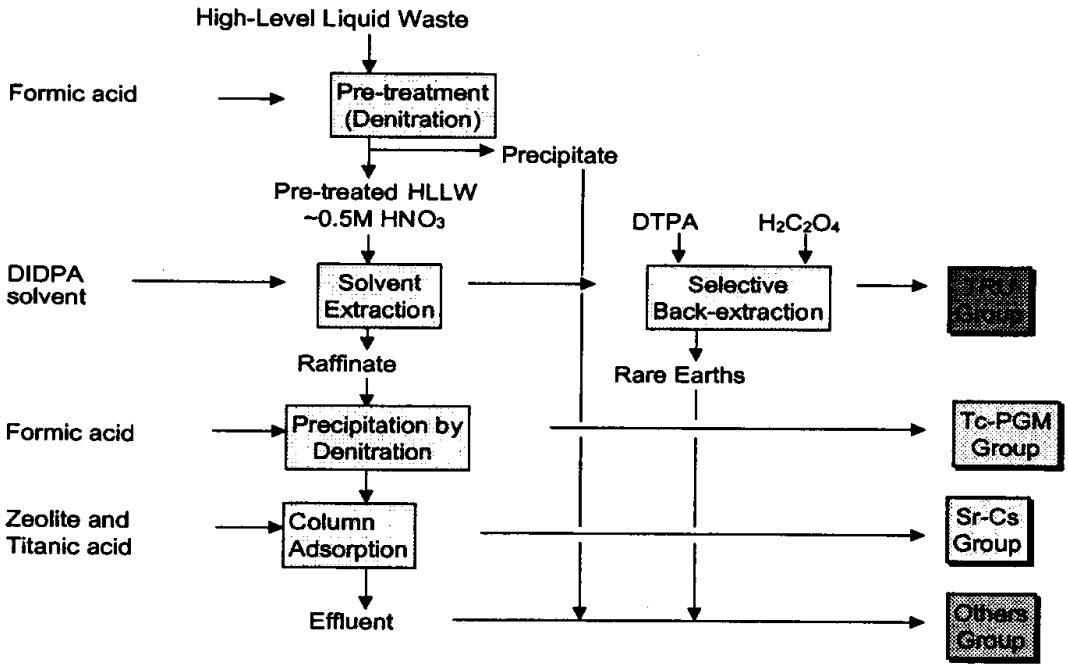
Figure 1. **Partitioning and transmutation system for long-lived radioactive nuclides at JAERI**




## 2. Partitioning process

A hot verification test of the 4-GPP with concentrated real HLLW was carried out in the Partitioning Test Facility in the hot cell [2]. For the preparation of the concentrated HLLW, about 14L (11 TBq) of the raffinate from the co-decontamination cycle of Purex Process were first denitrated and then concentrated to about 2.5L. The raffinate was obtained by two reprocessing tests with about 1 kg of spent fuel burned up to 8 000 MWd/t and with about 1.5 kg of spent fuel burned up to 31 300 MWd/t. The flow sheet is shown in Figure 2.

Figure 2. **Flow-sheet of 4-group partitioning process**




Results of the present test well agreed with the either result of previous tests using the unconcentrated real HLLW and the simulated HLLW added with a small amount of real HLLW. Table 1 summarises the fractional distribution of each element at the 1st mixer-settler. More than 99.998% of Am were extracted from the HLLW with the organic solvent containing 0.5M DIDPA – 0.1M TBP, and 99.986% of Am were back-extracted with 4M nitric acid. Cm showed the same behaviour as Am. Np and Pu were extracted simultaneously in a high yield, and more than 99.9% of them were back-extracted with oxalic acid. In the denitration step for the separation of Tc and PGM, pH of the solution was increased to 2.8 after the denitration, and then more than 90% of Rh and more than 97% of Pd were precipitated. About half of Ru were remained in the denitrated solution, but the remaining Ru were quantitatively precipitated after neutralization of the denitrated solution to pH 6.4, which was performed for the preparation of the feed solution to the adsorption step with the inorganic ion exchangers. In the adsorption step, both Sr and Cs were separated effectively. Decontamination factor for Cs was more than $10^6$ in all the effluent samples.

Table 1. **Fractional distribution (%) of each element at the 1st mixer-settler (from [2])**

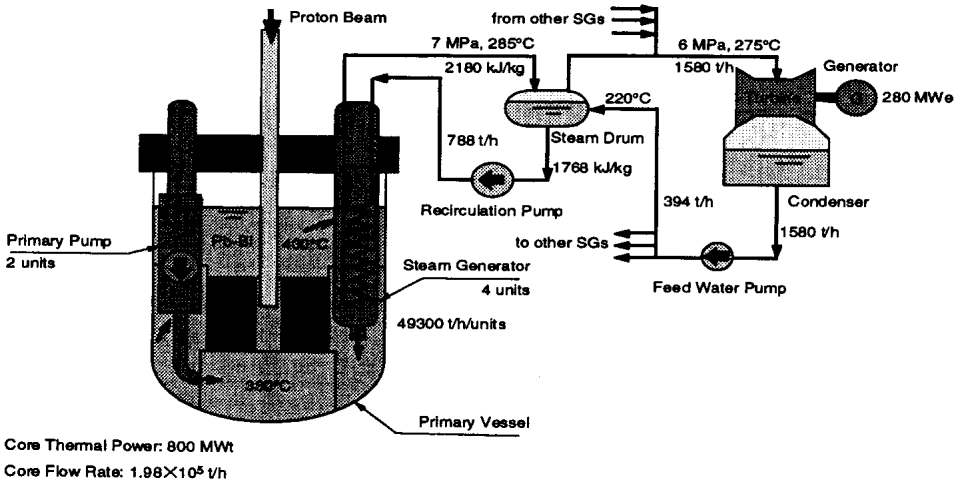
| **Element** | **Raffinate** | **Stripped with 4M $HNO_3$** | **Solvent** | **Mass balance** |
|---|---|---|---|---|
| Am | <0.002 | 99.986 | 0.012 | 77 |
| Cm | <0.001 | 99.984 | 0.015 | 79 |
| Eu | <0.0004 | 98.7 | 1.3 | 79 |
| Np | 1.8 | 0.3 | 97.9 | 110 |
| Cs | 99.95 | 0.05 | 0.001 | 106 |

The present 4-GPP necessitates a pre-treatment step, i.e. denitration-filtration, to reduce the acidity of an aqueous feed in harmony with the capability of diisodecylphosphoric acid (DIDPA). Thus as a modification effort of the 4-GPP, to eliminate the above step, more powerful ligand to extract actinides directly from high-level liquid waste (HLLW) has been developed in line with the CHON principle [3]. We found that a tridentate diglycolamide (DGA) with an ether oxygen at a centre of diamide molecule forms more stable complex with trivalent actinides and lanthanides than that of bidentate malonamides. Aiming at a high solubility of the extracted metal-DGA complex in n-dodecane solvent, to avoid the third-phase formation, and a high extractability toward all actinides, we have examined the effect of chain length of alkyl groups attached to the two amidic nitrogen atoms, and selected a ligand, tetraoctyl 3-oxapentandiamide (called TODGA hereafter) as the most proper DGA-extractant. To facilitate the development of TODGA-partitioning process, fundamental studies on i) the extraction behaviour of TODGA for various valency states of actinides, trivalent lanthanides, and some fission products ii) radiolytic degradation of TODGA by $^{60}$Co gamma rays have been carried out. The results of above investigation revealed that the TODGA is a satisfactory extractant to be applied to the process of separation of actinides and lanthanides (III) in the 4-GPP.

## 3. Transmutation process

Preliminary design of an 800-MWt lead-bismuth cooled accelerator-driven system has been developed as a dedicated transmutation system to be deployed into the second stratum in the double strata fuel cycle [4]. The system employs lead-bismuth for the target and primary coolant material. The plant has a pool type configuration and a power conversion system operated on a saturated steam cycle. An analysis of beam trip transient was made for this type of accelerator-driven transmutation plant [5]. Transients of the primary coolant temperature, the water/steam temperature, the water/steam pressure, the turbine flow rate, and the electric output were calculated using a simple network model based on a simplified flow diagram of ADS plant shown in Figure 3. The plant and turbine trips were required at 380 s after beam trip to prevent from overcooling. The maximum temperature swing was 185°C in lead-bismuth, and 82°C for in water/steam for the case when beam recovered at 370 s.

Figure 3. **Simplified flow model of ADS plant**




Mass balance in the proposed double strata transmutation system was analysed for cases where the type of power reactors in the first stratum is $UO_2$-LWR, MOX-LWR and FBR. The analysis shows that the transmutation rate for (Pu, MA) composition from a MOX-LWR becomes one half than that

from an $UO_2$-LWR. The number of transmutation systems and the amount of transmuted minor actinide are estimated for several possible scenarios of the future nuclear power development, assuming the deployment of transmutation systems starts in 2030. It was concluded that the introduction of ADS could play a significant role as "transmuter" in the back-end of fuel cycle [6].

A code system "ATRAS" was developed for the neutronics design of ADS [7]. The code system consists of the nucleon-meson transport code, Sn code and burn-up analysis code. In order to obtain the nuclear data required for the development of ADS, the Actinide File and the High Energy File were developed along with the JENDL General Purpose File.

## 4. Fuel process

Fabrication of MA nitride, irradiation tests of nitride fuel and the development of pyrochemical process for nitride fuel have been carried out [8].

### *4.1 Fuel fabrication process*

Fabrication of Pu and MA-bearing nitrides and preparation of the thermodynamic database have been carried out besides the irradiation tests of (U,Pu)N fuel up to 4.6 at%. High-purity AmN and (Pu,Cm)N were fabricated by carbothermic reduction of the dioxides by use of $^{243}$Am and $^{244}$Cm nuclides. X-ray diffraction patterns showed almost the single phase of NaCl-type structure. On the other hand, PuN pellets containing inert matrix nitrides such as ZrN and TiN were fabricated and characterised. Vapour pressure of Np(g) over NpN, (U,Np)N and (Np,Pu)N was measured by high-temperature mass spectrometry to clarify thermodynamic properties of the solid nitride phase. Thermodynamic property of Np(C,N), which is an intermediate product of carbothermic reduction for fabricating NpN from $NpO_2$, was also evaluated by both experiments and calculation. The results suggested that Np (C,N) could be treated as ideal solid solution as is the case of Pu (C,N). Measurements of heat capacity and thermal expansion of NpN and PuN are underway by use of the sintered sample for preventing oxidation. The irradiation of two $(U_{0.8}Pu_{0.2})N$ fuel pins at fast test reactor JOYO was completed in 1999 under the joint research with JNC. The non-destructive post irradiation examinations are underway and any failure of fuel pins was not observed. The destructive examinations will start in the latter half of this year.

### *4.2 Fuel reprocessing*

As for pyrochemical process, the electrochemical dissolution behaviour of NpN and PuN were measured by cyclic voltammetry and the equilibrium potentials of the nitrides in LiCl-KCl eutectic melt were determined. On the other hand, the electrochemical deposition behaviour of Pu at liquid Cd cathode was investigated. In this case the potential of deposition and dissolution shifted positively compared with the case of solid cathode in correspondence with a thermodynamic stabilisation by formation of intermetallic compound. Indeed, the formation of $PuCd_6$ phase was observed at the cathode by microprobe analysis. By adjusting electrochemical parameters such as current density during electrolysis, ten-gram scale of Pu was recovered at liquid Cd cathode with high Pu concentration. In addition, the electrochemical deposition behaviour of Np at liquid Cd cathode and the phase relationship of Am-Cd binary system were experimentally studied. Nitrogen releasing behaviour from NpN and PuN at an anode, and the results of distillation and nitrization of the Cd cathode after the electrolysis were examined. It was proved that the pyrochemical process is fundamentally suitable for recovery of expensive $^{15}$N-enriched nitrogen gas compared with the wet process.

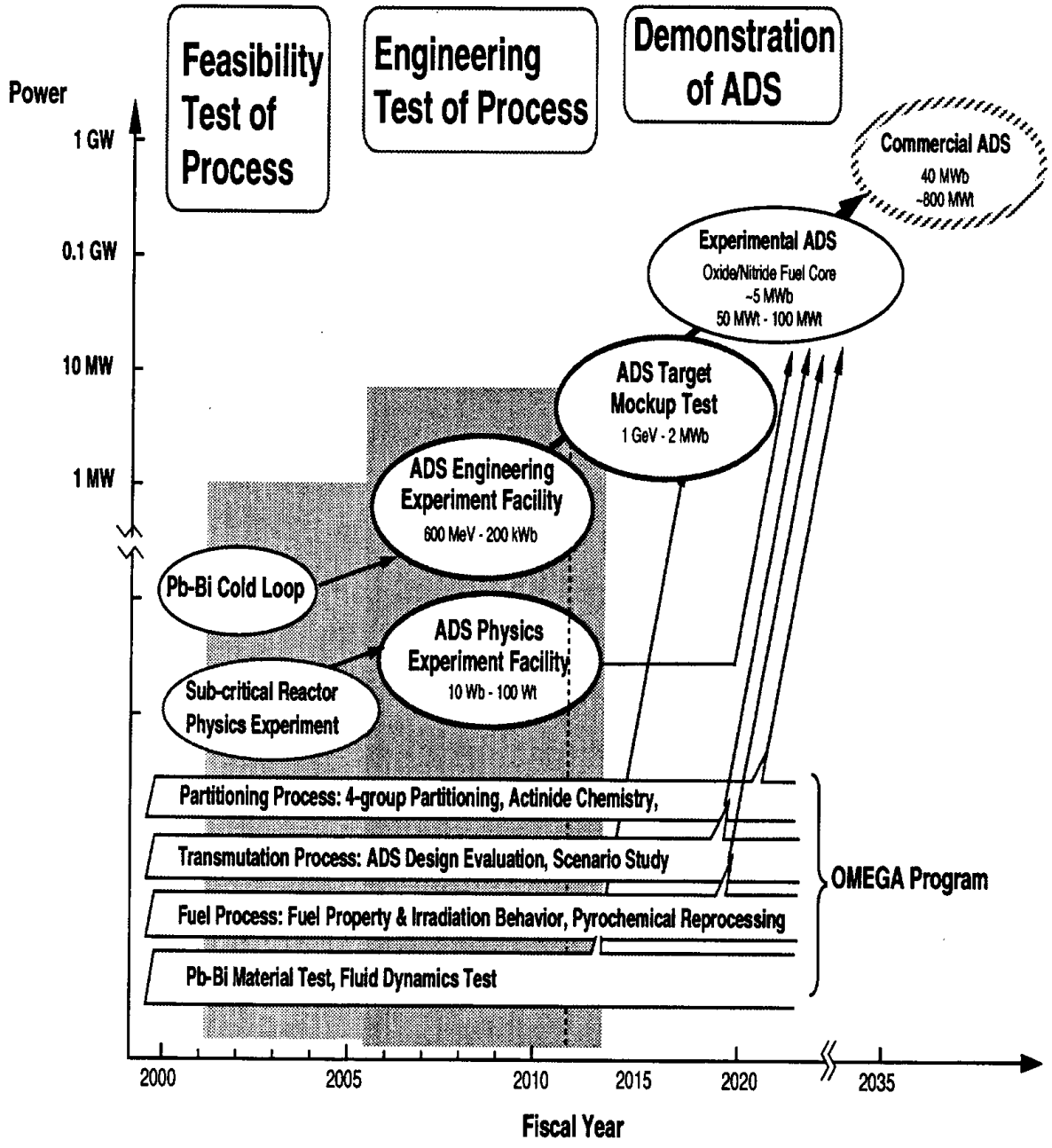
## 5. Concluding remarks

In 1999, the Atomic Energy Commission of Japan (AEC) established the Advisory Committee on Nuclear Fuel Cycle Back-end Policy to conduct the Check-and-Review of the outcome of the OMEGA programme. The Committee concluded, in the report issued in March 2000, that the present status of the programme would be the level of basic studies and tests, and that various concepts of the P&T system were evaluated and required technologies were developed. They also concluded that the future R&D should be proceeded in order to convert the high level waste into useful resources and to reduce the environmental impact associated with its disposal. Their recommended processes are as follows; to study the P&T implementation scenario taking account the situation of nuclear fuel cycle in Japan, to carry out basic experiments to demonstrate the feasibility of the process, and to conduct engineering scale experiments to obtain safety data of these systems.

After getting the results of the above mentioned C&R by AEC, JAERI will proceed the R&D in each process of the P&T system from the basic experimental step to the engineering mock-up step including the following areas Figure 4:

- **Pb/Bi material test**: One of the technical issues for ADS developments is the corrosion/erosion of material in liquid-bismuth coolant. The beam window represents a major technical challenge in ensuring the structural integrity as it suffers a high differential pressure load as well as thermal stress and radiation damage.
- **Am and Cm characteristics**: A “high temperature chemical cell” is to be constructed in Nuclear Fuel Cycle Safety Engineering Facility of JAERI for gram-scale experiments of Am and Cm.
- **ADS experimental facility**: Another issues to be tested or to be demonstrated are sub-critical reactor physics, and system operation and control. The experimental programmes to solve these technical issues for ADS developments have been planed within the framework of the JAERI-KEK Joint Project for High-Intensity Proton Accelerators.

Figure 4. **Scenario for development of ADS**



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