

STUDIES ON ACCELERATOR-DRIVEN TRANSMUTATION SYSTEMS

T. Takizuka, T. Sasa, K. Tsujimoto, and H. Takano

Japan Atomic Energy Research Institute

Tokai-mura, Ibaraki-ken, 319-1195,

Japan

Abstract

Research and development on transmutation of long-lived radioactive nuclides are being carried out with an emphasis placed on the dedicated accelerator-driven systems at the Japan Atomic Energy Research Institute (JAERI) under the Japanese long-term program for research and development on partitioning and transmutation technology (OMEGA Programme). The preliminary design of the sodium-cooled solid system has been developed as a reference system based on the current LMFBR technology. Concurrently with the study on the sodium-cooled system, we made a preliminary design study of lead-bismuth cooled system as an option. Transient responses of temperatures and stresses due to accelerator beam trips were evaluated for the fuel pin and the beam window of the proposed experimental facilities

Introduction

Research and development on transmutation of long-lived radioactive nuclides are being carried out with an emphasis placed on dedicated accelerator-driven systems at the Japan Atomic Energy Research Institute (JAERI) under the Japanese long-term program for research and development on partitioning and transmutation technology (OMEGA Program). Design studies are being made for accelerator-driven transmutation systems (ADTSs). Design of the current reference ADTS follows that of contemporary sodium cooled fast breeder reactors (FBRs). The major reasons to chose sodium as coolant are its excellent thermal performance and technology maturity. However, a preliminary design study was recently started for a heavy liquid-metal cooling option of ADTS at JAERI. Monju (714-MWt sodium cooled prototype fast reactor) suffered a leakage and fire of secondary sodium in December 1995. This incident has caused the people to feel concern about safety of sodium coolant technology and stimulated much interest in alternative coolant options, such as lead, lead-bismuth, and He, for fast reactors. The main purposes of the design study of heavy liquid-metal cooling option are to determine the performance, to assess the feasibility and to identify the technical issues of a heavy liquid-metal cooled design in comparison with the sodium cooled reference design.

The ADTS was specially designed to transmute minor actinides (MAs) from about 10 units of 3000-MWt light water reactor (LWR) in a hard neutron energy spectrum and a high neutron flux. Such dedicated transmuter can be very efficient and effective for MA transmutation. In this context, JAERI has been pursuing the strategy of transmutation with dedicated transmuters, rather than recycling to commercial power reactors and proposing a concept of a double-strata fuel cycle consisting of a power reactor fuel cycle (the first stratum) and a P-T cycle (the second stratum) [1,2]. In this scenario, a reprocessing plant, a partitioning plant and a dedicated transmuter will be co-located in one site.

The experimental program for development and demonstration of ADTS technology is being planned under the JAERI Neutron Science Project. Pre-conceptual design study is being made for the experimental facilities (30 – 60 MWt experimental system and high-power target experimental facility). Large fluctuations and frequent trips of the incident proton beam may be inevitable in these facilities. These changes in beam intensity will cause changes in temperatures and stresses in reactor components. This will introduce thermal stress problems related specifically to accelerator-driven system. To assess the impact on the structural integrity and lifetime of the components, transient responses of temperatures and stresses were evaluated for the fuel pin and the beam window of the experimental system.

Conceptual design study of transmutation systems

The concepts of ADTS have been developed at JAERI. The ADTS is specially designed for transmutation purpose as a dedicated transmuter to be deployed in the P&T cycle of the double-strata fuel cycle. In the proposed ADTS, an actinide loaded subcritical core is driven by a high-intensity proton accelerator with several tens of MW beam and uses fast neutrons for efficient and effective transmutation of actinides. The current design aims at supporting about 10 units of large-scale LWRs with 1000-MWe capacity.

The preliminary design of the sodium-cooled solid system has been developed as a reference system based on the current sodium cooled FBR technology. The reference system employs solid tungsten for the spallation target and MA mononitride for fuel of subcritical core. Nitride is adopted as the fuel material because of its excellent thermal property. The other advantage of nitride fuel is that it can be processed with the pyrochemical reprocessing, [3] and, hence, the fuel cycle facilities can be very compact and cost effective.

Recently, JAERI started a design study of a heavy liquid-metal cooled ADTS as an alternative possible option to re-evaluate its technical viability. Although the heat transfer capability of lead or lead-bismuth is inferior to that of sodium, it has several advantages as coolant for accelerator-driven transmutation systems. Liquid lead or lead-bismuth is particularly suited to the target material, this eliminating the need for a distinctive solid target. It also offers the possibilities to achieve a harder neutron energy spectrum and to avoid a positive void reactivity coefficient. Intermediate heat exchangers and secondary heat transport loops will possibly be eliminated with lead or lead-bismuth coolant. Lead-bismuth eutectic coolant offers much lower system operating temperatures than lead coolant. The lower operating temperature will alleviate the severe problems of material corrosion/erosion in heavy metal coolants.

A computer code system ATRAS has been developed for the design of accelerator-driven transmutation system. [4] The cascade code NMTC/JAERI [5] simulates the proton-induced nuclear spallation, subsequent internuclear transport process for energies above 20 MeV. It also calculates high-energy fission reaction as a competing process with evaporation. Neutronic calculation below 20 MeV is carried out using transport codes, TWODANT and MCNP4A. The time evolution process of transmutation products is calculated by SPCHAIN code and by BURNER code for energies above and below 20 MeV, respectively.

Sodium cooled accelerator-driven transmutation system

The design of the sodium cooled solid system is based on a sodium cooled FBR. Proton beam is injected through a beam window into the tungsten target at the center of the target/core. The subcritical core loaded with actinide nitride fuel surrounds the target. The target consists of multiple layers of tungsten disk with through holes for coolant passage. The target is designed to maximise the neutron yield and to flatten the axial power distribution. The target and fuel subassemblies are cooled by forced upward flow of primary Sodium. Impinging flow of coolant from the target exit cools the beam window. The concept of the sodium cooled solid system is shown in Figure 1.

Figure 1 Sodium cooled ADTS concept

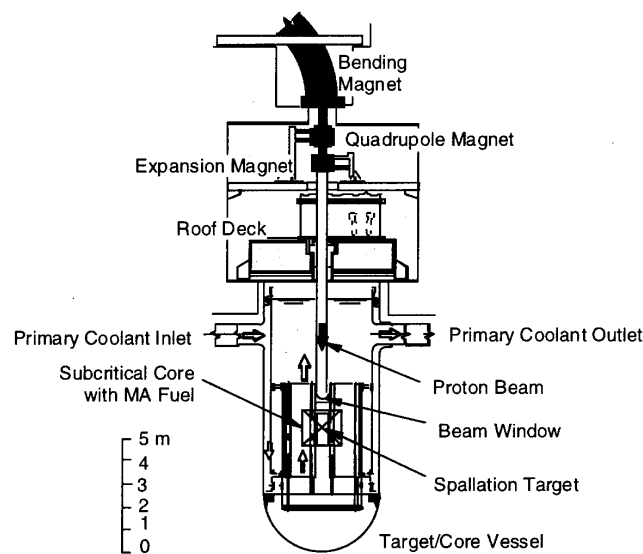
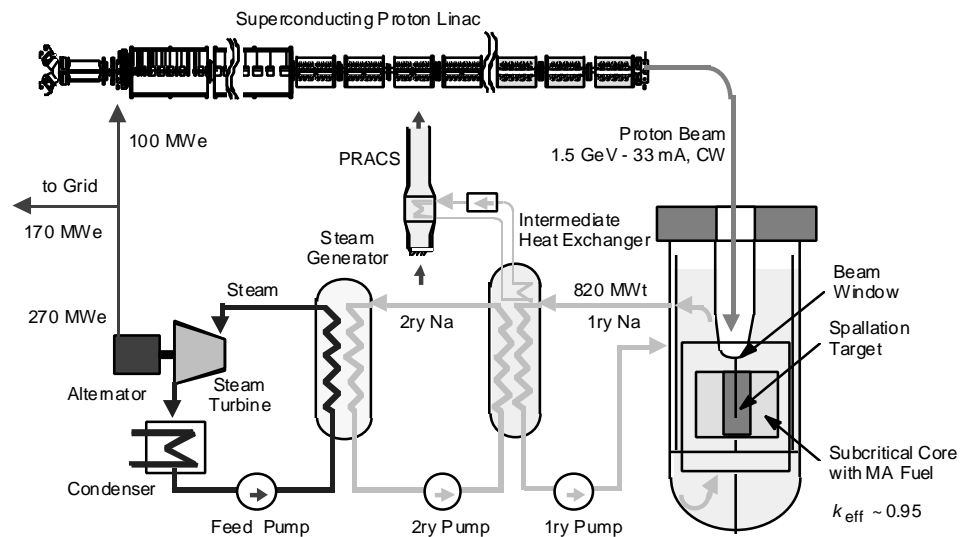


Table 1 Major parameters of the sodium cooled ADTS

| | |
|--------------------------------------|---------------------------------------|
| MA/Pu Inventory | 1950/1300 kg |
| k_{eff} (Initial/Max./Min.) | 0.93/0.94/0.92 |
| Sodium Void Coefficient | +4.5% dk/k |
| Doppler Coefficient | $-2.2 \times 10^{-4} \text{ T dk/dT}$ |
| Thermal Power | 820 MW |
| Transmutation Rate | 250 kg/y |
| Power Density (Max./Ave.) | 550/380 MW/m ³ |
| Coolant Temperature (In/Out) | 330/430 °C |
| Coolant Velocity (Max.) | 8 m/s |

With a 1.5 GeV - 33 mA incident proton beam, the target/core having an effective neutron multiplication factor of around 0.95 produces 820-MW thermal power. The net MA transmutation rate is approximately 10%/y, at a plant load factor of 80%. Heat transport and power conversion systems in the plant design are similar to those for a sodium cooled FBR plant. Electricity of 270 MW is generated through conventional steam turbine. One third of electric power is supplied to its own accelerator operation. The major parameters and the schematic diagram of the ADTS plant are shown in Table 1 and Figure 2, respectively.

Figure 2 Schematic diagram of the sodium cooled ADTS plant



Lead-Bismuth cooled accelerator-driven transmutation system

There are several advantages of heavy liquid metal coolants (lead or lead-bismuth) for fast neutron systems in comparison with sodium coolant, though their thermal property is inferior to that of sodium. In accelerator transmutation systems, lead or lead-bismuth can play roles of both coolant and spallation target material. Their neutron slowing down power is smaller than that of sodium, and hence neutron spectrum becomes harder in the lead or lead-bismuth cooled core. The hard spectrum is preferable for MA transmutation. Their chemical inertness is particularly attractive for safety. This also offers the possibility to eliminate secondary heat transport loops.

Corrosion which is one of the most important problems in lead or lead-bismuth cooled systems is significant at high temperatures. The melting point of lead-bismuth is almost the same as that of sodium while that of lead is considerably higher. So, we selected the lead-bismuth as the coolant material for the dedicated ADTS.

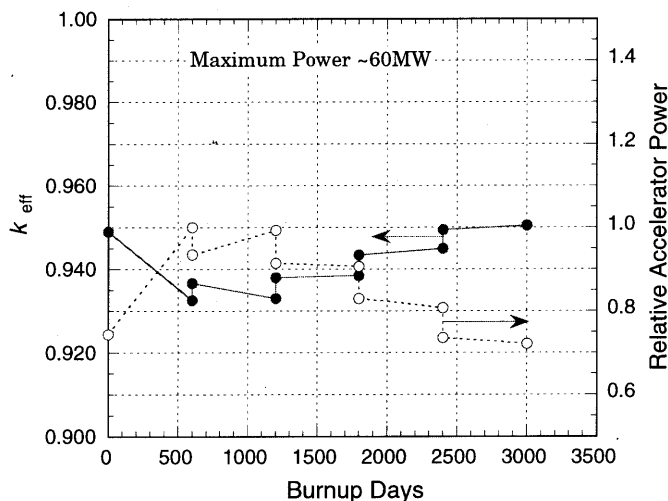
In this study, we investigated the basic characteristics of lead-bismuth cooled ADTS, such as the change of the multiplication factor, the fuel composition change and the transmutation rate. The MA inventory and the transmutation rate are especially important factors in the transmutation system. It is also significant to suppress the excess reactivity change during burnup for minimising the change of the proton beam current.

Calculation of neutronic characteristics

The survey calculation was performed to investigate the neutronic characteristics of lead-bismuth cooled ADTS. First, the fuel compositions were determined by adjusting the Pu content and the fraction of inert matrix to set the initial subcritical level at 0.95. Second, the burnup calculations were done for these fuel compositions. The MA transmutation rate and the burnup swing are especially important to estimate the ADTS performance. The purpose of the survey was to optimise the core parameters for maximising the MA transmutation rate and minimizing the burnup swing. The quantitative goals were the MA inventory below 2500 kg, the MA transmutation rate above 10%/y, and the burnup swing below 2% during 10 years full power operation.

The core sizes were determined by heat-balance calculation using the fuel pin pitch to diameter ratio (P/D) and coolant velocity as the parameters. The P/D value was varied in the range from 1.5 to 1.9. The coolant velocity was varied from 1.2 to 2.0 m/s. In this calculation, the core thermal power was assumed to be 800 MW. The coolant temperature rise through the core was set to 100 °C, and the average linear power, 300 W/cm. The core height was fixed at 100 cm and the diameter of the spallation target region was set at 50 cm, in all cases.

Figure 3 Changes of the multiplication factor and proton beam power with burnup



Fuel is mixture of mono-nitride of Pu and MA and inert matrix (ZrN). The initial Pu content was varied from 0 to 60%. ¹⁵N enriched nitrogen are used for both (Pu,MA)N and ZrN. The compositions of Pu and MA used in this study are those in the spent fuel of the 33 GWd/t burnup in PWR after 5 years cooling.

The multiplication factor was calculated by the diffusion calculation. The group constants of fuel region were obtained by the collision probability method considering heterogeneous effect by the SLAROM code. [6] The cross section library used in SLAROM was JFS-3 type [7] 70 group constants set based on the JENDL-3.2 library. [8] The diffusion calculation was done by the modified CITATION code [8].

The burnup calculation was done by the ABC-SC code system. The ABC-SC code system consists of SLAROM, CITATION and ORIGEN2. [10] The changes of effective cross section with burnup for about 40 actinide isotopes including the JENDL-3.2.2 library were considered in each burnup step in ABC-SC. For other isotopes, the values in ORIGEN's fast reactor library were used. The burnup calculations were done for 5 burnup cycles. We assumed one burnup cycle consists of 2 years burnup and 3 years cooling. After the cooling, FPs were removed and MA of equal mass to the FPs was added to the burnup fuel to recycle.

Figure 3 shows the changes of the multiplication factor and the proton beam power for the ADTS with optimum set of core parameters. In this case, the initial Pu fraction is 40% and P/D is 1.5. The coolant velocity is 2.0 m/s, corresponding to the power density of about 180 W/cm³. The figure indicates that the change of proton beam power is about 30%, though the burnup swing in 10 years operation is only 1.8%. The maximum beam power is needed at the maximum system subcritical level for constant power operation, corresponding to about 60 MWb.

The major core parameters of the lead-bismuth cooled ADTS are shown in Table 2.

Table 2 Major parameters of the lead-bismuth cooled ADTS

| | |
|--------------------------------------|---|
| Core Thermal Power | 800 MW |
| Core Height | 1000 mm |
| Core Radius | 1200 mm |
| k_{eff} (Initial/Max./Min.) | 0.95/0.95/0.94 |
| Linear Power Rating (Max./Ave.) | 520/300 W/cm |
| Power Density (Max./Ave.) | 310/180 MW/m ³ |
| MA/Pu Inventory | 2500/1660 kg |
| Fuel | (MA, Pu)N, inert matrix ZrN initial Pu fraction 40% N-15 enriched |
| Pin Pitch-to-Diameter ratio | 1.5 |
| Coolant Temperature (In/Out) | 330/430 °C |
| Coolant Velocity (Max.) | 2 m/s |
| Sodium Void Coefficient | -4.8% dk/k |
| Doppler Coefficient | $-3.7 \times 10^{-4} \text{ T dk/dT}$ |
| Burnup Swing | 1.8 % |
| Variation in Beam Power | 30 % |
| MA Transmutation Rate | 500 kg/cycle (20%/cycle) |

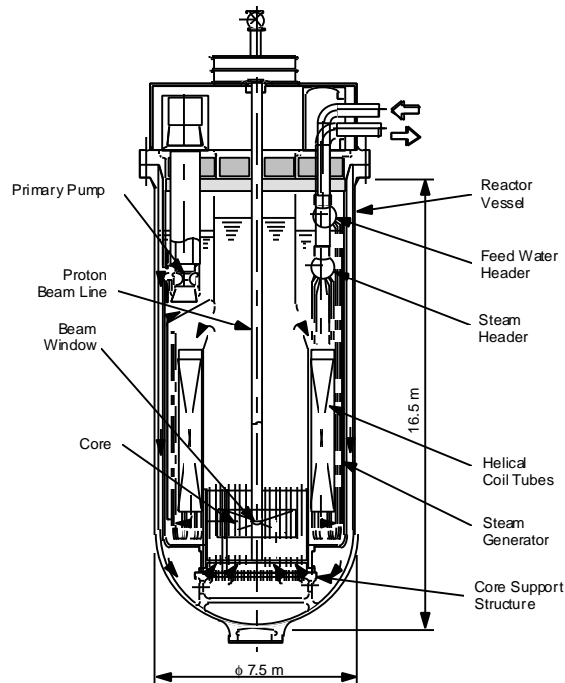
Conceptual design of lead-Bismuth cooled ADTS plant

The plant design was principally based on the early design study of a 1500-MWt lead cooled fast reactor at JAERI. [11] For designing lead-bismuth cooled system, it is necessary to overcome the subject of aseismatic design and thermal stress resistance because the reactor will be excessively

massive. The loop type configuration, which was employed in the sodium cooled ADTS design, is not suitable for lead-bismuth cooled system, because it will be difficult to realise the heavy piping system. So, the pool type reactor is selected for the lead-bismuth cooled ADTS. Intermediate heat transport system is possible to be eliminated with lead-bismuth coolant. Cr-Mo steel is used as the structural material considering the corrosion resistance in lead-bismuth, mechanical strength and ductility. The plant heat balance was calculated and the sizes of the primary pumps, the steam generators and the core vessel were determined.

The concept of the lead-bismuth cooled ADTS is shown in Fig. 4. The core, the core support structure and the primary heat transport system components are contained in the reactor vessel. The primary heat exchangers are the integral type steam generators consisting of helical coil tubes. This type of steam generator allows to reduce the reactor vessel size. The space between the outer shell of steam generator and the reactor vessel is the cold-leg flow path of primary coolant. The primary pumps are located at the cold region of the primary coolant flow path. The pumps are located above the helical coil steam generator, and the steam generators are suspended from the reactor upper flange from the viewpoint of maintenance. The plant has the water-steam cooled decay heat removal system, that is the primary reactor auxiliary cooling system (PRACS). The PRACS heat exchanger coils are located above the steam generator coils. Electrical heaters are wrapped around the PRACS piping to prevent lead-bismuth from freezing.

Figure 4. Concept of the lead-Bismuth cooled solid ADTS



The present study is preliminary one, and there remain many technical issues to be solved before finalising the design. One of the major technical issues is corrosion/erosion of material in lead-bismuth coolant. Candidate materials are 2.25Cr-1Mo steel, 9Cr-1Mo steel, etc. The dedicated ADTS does not have the primary purpose of power generation. It can be operated at about 100 °C lower temperatures than commercial sodium cooled fast reactors, sacrificing the efficiency of thermal to electricity power conversion. The low operating temperature is expected to mitigate the corrosion/erosion problem.

The beam window suffers from the stress due to the differential pressure between the accelerator vacuum and system operating pressure. In the lead-bismuth cooled system, the differential pressure is much larger due to high density of lead-bismuth than in the sodium cooled system. The pressure loads together with thermal stress cause a very difficult problem of beam window mechanical strength. This problem could further worsen in a high flux environment of protons and neutrons.

In lead-bismuth coolant, fuel subassemblies are subjected to upward buoyancy force that is larger than downward gravitational force. Design measure for mechanical constraint should be needed to keep the subassemblies fixed.

The problem of ^{210}Po generation by capture reaction of ^{209}Bi arises in lead-bismuth cooled system. The amount of radioactivity in the primary coolant was estimated to be the same level as conventional fast reactors. The estimation of radioactivity and toxicity of the spallation products is now underway. Some purification system to remove ^{210}Po and spallation products from primary coolant will be necessary.

Effects of accelerator beam trips on fuel pin and beam window

One of the most important effects of beam trips is thermal fatigue of ADS component materials. A beam trip changes temperatures in ADS components. Changes in temperatures causes changes in thermal stresses and thermal strains in the components. Repeated temperature changes (thermal cycling) due to frequent beam trips can cause thermal fatigue of the component materials. Damages caused by thermal fatigue can lead to degradation of their structural integrity and reduction of their lifetime. Rather frequent replacement of the components of concern and/or special design measures to mitigate the problem might be required, if the accelerator reliability could not be sufficiently improved.

Preliminary analyses were made on thermal and structural responses of a fuel pin and a beam window for the experimental facilities [12] planned under the Neutron Science Project.

Analyses of fuel pin

Two-dimensional thermal-hydraulic and structural analyses were made on the single fuel pin with the average power of the planned 60 MWt experimental system. Conditions of analyses are listed in Table 3.

The maximum fuel temperature calculated is $1\,520^\circ\text{C}$, sufficiently lower than the melting point around $2\,800^\circ\text{C}$. Tensile strength of the oxide fuel pellet is around 10 kg/mm^2 at $1\,500^\circ\text{C}$. Calculated maximum tensile stress of the fuel pellet is around 37 kg/mm^2 . It is expected that one or two radial cracks will be formed. The cracks will relieve the stress and preventing from further crack formation and propagation.

It is predicted there are no possibilities of tensile/creep rupture of the 316 SS cladding, since the internal pressure of fuel pin due to gaseous FP release is low. With an appropriate gap, fuel clad mechanical interaction can not occur. The major factor that can affect the integrity of cladding is fatigue caused by thermal cycling.

The maximum thermal strain of the cladding is 2.3×10^{-4} . Since the design fatigue strain range at 10^7 cycles is 10^{-3} at 430°C , it is evaluated that fatigue damage to cladding is negligible.

Table 3 Conditions of analyses of fuel pin

| | |
|------------------------------|---------------------------------------|
| Fuel | UO ₂ |
| Bond | Helium |
| Cladding | 316 SS |
| Pin Diameter | 5.4 mm |
| Pin Pitch | 7 mm |
| Active Core Height | 850 mm |
| Coolant | Sodium |
| Coolant Temperature (In/Out) | 330/430°C (normal operation, average) |
| Linear Power Rating | 120 W/cm (normal operation, average) |

Analyses of beam window

Two-dimensional thermal-hydraulic and structural analyses were made on the beam window of the planned high-power target experimental facility. The beam window is a hemispherical cap made of 316 SS, having a radius of 120 mm and a thickness of 1 mm. The incident proton beam with a power of 7 MW has a diameter of 200 mm and a uniform current density distribution. The beam window is cooled by upward flow of sodium coolant. The operating pressure of the sodium coolant at the level of the beam window is 3 Mpa.

The maximum operating temperature calculated is 525°C, and the temperatures at evaluated cross sections are in the range from 340 to 515°C. Results were evaluated for a high-temperature operating time of 8 760 h according to “High-temperature Design Guideline for FBR Prototype Class 1 Components” (STA) and “Technical Standard for Structures of Nuclear Facilities for Power Generation” (MITI).

All stresses and strains are well within their allowable limits, and the allowable number of operating cycles is greater than 10⁶. Fatigue damage to the beam window was evaluated to be negligible.

Concluding remarks

A preliminary design study was made for a lead-bismuth cooled accelerator-driven system as an possible alternative option to the sodium cooled solid system concept based on the current LMFBR technology. The core parameters were determined to achieve minimum variation in the effective multiplication factor during burnup cycles. Pool-type configuration was selected for the lead-bismuth cooled system from the point of aseismatic design view rather than loop-type used for the sodium cooled reference system. Major technical problems to be resolved are material compatibility, beam window design, etc.

Preliminary analyses were performed on thermal and structural responses of a fuel pin and a beam window during a normal operation and a beam trip transient. It was estimated that fatigue damages to the fuel cladding and the beam window caused by beam trips are negligible. More detail design and further analyses are needed to formulate design considerations, R&D needs, and the requirements of accelerator reliability.

REFERENCES

- [1] H. Murata and T. Mukaiyama, *Atomkern-Energie Kerntechnik*, 45, p. 23 (1984).
- [2] T. Mukaiyama, *Importance of the double strata fuel cycle for minor actinide transmutation*, Proceedings 3rd OECD/NEA Int. Information Exchange Mtg. on P-T, p. 30 (Cadarache, 1994).
- [3] T. Ogawa *et al.*, *Nitride Fuel Cycles on Pyrochemistry*, Proceedings. Int. Conf. Future Nuclear Systems, “Global ’97” (Yokohama, 1997).
- [4] T. Sasa *et al.*, *Conceptual Design Study and Code Development for Accelerator-Based Transmutation System*”, Proceedings Int. Conf. Future Nuclear Systems, “Global ’97”, (Yokohama, 1997).
- [5] Y. Nakahara *et al.*, “NMTC/JAERI – A Simulation Code System for High Energy Nuclear Reactions and Nucleon-Meson Transport Process, JAERI-M 82-198 (in Japanese) (1982).
- [6] M. Nakagawa and K. Tsuchihashi, “SLAROM : A Code for Cell Homogenization Calculation of Fast Reactor”, JAERI 1294 (1984).
- [7] H. Takano and Y. Ishiguro, *Production and Benchmark Tests of Fast Reactor Group Constant Set JFS-3~J2*, JAERI-M 82-135 (1982).
- [8] T. Nakagawa, *et al.*, *Japanese Evaluated Nuclear Data Library Version-3 Revision-2 : JENDL-3.2*, J. Nucl. Sci. and Technol., 32, 1259 (1995).
- [9] T. B. Fowler, D. R. Vondy and G. W. Cunningham, *Nuclear Reactor Core Analysis Code : CITATION*, ORNL-TM-2496 (1969).
- [10] A. G. Groff, “ORIGEN-2 : A Revised and Updated Version of the Oak Ridge Isotope Generation and Development Code”, ORNL-5621 (1980).
- [11] H. Takano *et al.*, *A Design Study for Inherent Safety Core, Aseismicity and Heat Transport System in Lead-Cooled Nitride-Fuel Fast Reactor*, Proceedings ARAA' 94, Pittsburgh, Pennsylvania, April 17-21, 1994, vol. 1 p.549-556 (1994).
- [12] T. Takizuka, *et al.*, *Accelerator-Driven Transmutation System Demonstration Experiments at JAERI*, Proceedings Int. Conf. Future Nuclear Systems, “Global ’97”, (Yokohama, 1997).