# R&D ACTIVITIES FOR PARTITIONING AND TRANSMUTATION IN KOREA

Jae-Hyung Yoo and Tae-Young Song
Korea Atomic Energy Research Institute
P.O. Box 105, Yusong, Daejeon, 305-600 KOREA

## ABSTRACT

According to the Korean long-term plan for nuclear technology development, KAERI is conducting a few R&D projects related to the proliferation-resistant back-end fuel cycle. The R&D activities for the back-end fuel cycle are reviewed in this work, especially focusing on the study of the partitioning and transmutation(P&T) of long-lived radionuclides. The P&T study is currently being carried out in order to develop key technologies in the areas of partitioning and transmutation. The partitioning study is based on the development of pyroprocessing such as electrorefining and electrowinning because they can be adopted as proliferation-resistant technologies in the fuel cycle. In this study, various behaviors of the electrodeposition of uranium and rare earth elements in the LiCl-KCl electrorefining system have been examined through fundamental experimental work. As for the transmutation system, KAERI is studying the HYPER (HYbrid Power Extraction Reactor), a kind of subcritical reactor which will be connected with a proton accelerator. Up to now, a conceptual study has been carried out for the major elemental systems of the subcritical reactor such as core, transuranic fuel, long-lived fission product target, and the Pb-Bi cooling system, etc. In order to enhance the transmutation efficiency of the transuranic elements as well as to

strengthen the reactor safety, the reactor core was optimized by determining its most suitable subcriticality, the ratio of height/diameter, and by introducing the concepts of optimum core configuration with a transuranic enrichment as well as a scattered reloading of the fuel assemblies.

## INTRODUCTION

The nuclear power industry in Korea has grown dramatically since the first commercial nuclear power plant, Kori #1, started operation in 1978. Sixteen nuclear power plants (14 PWRs and 4 PHWRs) are currently in operation, supplying about 40% of total electricity demand in Korea. As of the end of 2002, the accumulated spent fuel accounts for about 6,000 ton that is stored at four reactor sites. The cumulative amount is prospected to reach 11,000 ton by 2010. However, the Korean government has not decided on any definite policy yet for back-end fuel cycle, while sticking to the policy of "wait & see". In the field of R&D only, a few optional studies, such as DUPIC (Direct Use of PWR spent fuel In CANDU), transmutation, and direct disposal, are being carried out in order to find an effective solution for the long-term management of spent fuels. According to the long-term plan of nuclear technology development, KAERI is conducting an R&D project of transmutation with the objective of key technology development by 2006 in the areas of partitioning and the transmutation system. As for the transmutation system, the HYPER (HYbrid Power Extraction Reactor) system combined by a proton accelerator and a subcritical reactor is being considered as an appropriate means for the Korean situation trying to place the priority of back-end fuel cycle on the non-proliferation of nuclear fissile materials. The subcritical reactor is supposed to have a fast neutron flux composed of spallation neutrons that are created by the hitting of accelerated proton beams against the spallation target. This system, therefore, is anticipated to tide over a certain extent of impurities (fission products) left in the fuel at the time of transuranic element recycle. It will certainly decrease the burden of partitioning of transuranic elements and also contribute to the requirement of non-proliferation of the P-T cycle because it is not necessary to further purify the fuel material after partitioning. The long-lived fission products such as Tc-99 and I-129 can also be loaded in the system as irradiation targets and then transmuted by neutron capture, which would be achievable by local moderation of the fast neutrons. In parallel with this, the partitioning study is focused on the development of pyroprocessing technology based on the electrolysis of molten salts. The basic study currently being conducted at KAERI includes some experimental tests of lab-scale electrorefining and electrowinning by employing only non-radioactive materials. This study is aiming at analyzing its technological applicability to the P-T cycle and also its advantages in the long-term management of spent fuels in the viewpoint of disposal as well as monitoring of the environment surrounding the repositories. The experimental work employing transuranic compounds will be done in the future on the basis of international collaboration programs.

## STRATEGY OF PARTITIONING AND TRANSMUTATION

The nuclear transmutation, though it is not fully developed yet for commercialization, is taken into account as a proper option for the solution of future spent fuel management. It may be an efficient way to relieve the risks of radiological contamination of the environment caused by probable release of long-lived radionuclides from the repositories. Since transmutation accompanies P-T(Partitioning & Transmutation)cycle where long-lived radionuclides must be partitioned and recycled, partitioning is an essential ingredient to complete the transmutation technology. Recently, pyrochemical separation method, though it is still in the R&D stage, is attracting a great interest as a prospective partitioning of long-lived radionuclides because it has noticeable advantages over wet processes, especially in terms of proliferation resistance as well as economy. According to KAERI's long-term R&D program, we are supposed to develop transmutation technology based on a hybrid transmutation system as well as pyrochemical technology as the basis of the P-T cycle.

Conversion of oxide fuel material is being taken into account in order to reduce the oxides into metallic forms by using lithium in a lithium chloride bath. The materials of metallic forms then will be treated in the electrorefining process [1]. The concept of two step-electrorefining employing a dual cathode system is being investigated. The first step will be aimed at recovering only uranium on the solid cathode at a certain value of electrical potential, while the second step aimed at recovering all the remaining actinides together into the liquid cathode at a newly adjusted potential. The molten salt waste from the electrorefining should be solidified as a final waste form to be disposed of. The treated waste will then be studied to solidify into ceramic form, which has very low leaching of radionuclides.

The objective materials to be transmuted are the group of transuranic elements most of which are composed of long-lived radionuclides, and the long-lived fission products such as I-129 and Tc-99 which also have long half-lives. It is necessary to separate these radionuclides from the wastes before loading them into a transmutation system because it is impossible to transmute only the long-lived radionuclides selectively.

The basic concept of recent transmutation system, is focused on the gaining of the double advantage by the use of transuranic elements as a nuclear fuel material. The advantages are the consumption of transuranic elements as well as the generation of electricity by using the transuranic elements. Based on this concept, some technological requirements are needed for the partitioning as well as for the transmutation system. The first requirement is the exclusion of uranium as a fuel source in the transmutation system because it can generate new transuranic elements by neutron capture, building up new transuranic elements in the fuel. The consumption rate of transuranic elements, however, will not reach 100% in one life-cycle, requiring another partitioning in order to separate long-lived radionuclides and then recycle them again into the transmutation system, which is called P-T cycle (see Fig. 1). Another requirement for the P-T cycle is that the partitioning should be done under the condition of non-proliferation and higher economy. Transuranic elements, therefore, are required to be separated as a mixture which is directly used as a fuel material in the transmutation system without any more partitioning. This requirement may be satisfied by the introduction of the pyroelectrochemical separation even though it is not at a commercial stage yet.

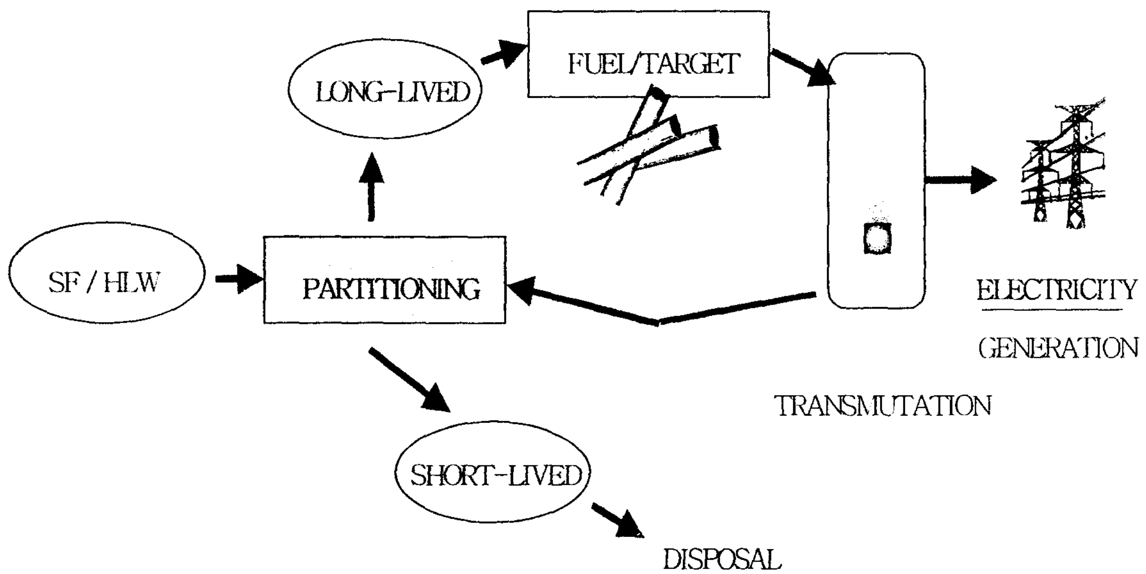


**Figure 1. A concept of partitioning and transmutation cycle**

The technological requirements which are mentioned above can be summarized as follows :

i ) Exclusive use of uranium as a fuel source in the transmutation system

ii ) Partitioning based on nonproliferation

iii) Higher rate of transmutation and thus less number of recycle of long-lived radionuclides.

However, a great expenditure would be needed for the partitioning and recycling of long-lived radionuclides, and also for the operation of the transmutation system, causing the economy of the P-T cycle to be negative. On the contrary, there are also some benefits that may turn the economic barometer from the negative to the positive direction. They would come from the electricity generated from the transuranic elements as well as the cost-saving in high-level waste disposal. If the economic analysis is extended to the environmental friendlyness, it may bring additional positive effect also.

## PARTITIONING BY PYROPROCESSING

### Recovery of uranium

Uranium is contained in spent fuels or high-level wastes from reprocessing plants as a major and a minor component, respectively. As described before, since uranium is not so desirable as a fuel source of the transmutation system, it is necessary to remove it from the spent fuels or high-level wastes in advance in the partitioning step. The fundamental studies of pyroprocessing conducted in KAERI in order to remove uranium are introduced below.

### Electrorefining

Electrorefining is a key technology to recover uranium and transuranic elements from spent fuels or high-level radioactive wastes [2,3]. Since each metal chloride/fluoride has a unique value of Gibbs free energy of formation in the molten salt electrolysis [4], uranium can be selectively reduced and deposited on the surface of cathode by the adjustment of the electrical potential between anode and cathode. Consequently, uranium can be recovered as an electrodeposition at the cathode.

Fig. 2 shows the schematic diagram of experimental electrorefining system. The electrolysis cell is installed in a furnace, which is located under the bottom of the glove box. The electrolysis cell and glove box are filled with an inert gas in order to protect the molten salt from being reacted undesirably with oxygen or moisture. The offgas from the electrolysis cell is treated with alkali solution and water so that chlorine, hydrochloride, that could be formed in the cell, or some evaporated compounds can be trapped in the liquids before going to the ventilation system.

Fig. 3 shows an experimental result of uranium deposition on the solid cathode, in which the molten salt LiCl-KCl was used as an electrolyte at 1.3 V and 500 °C.

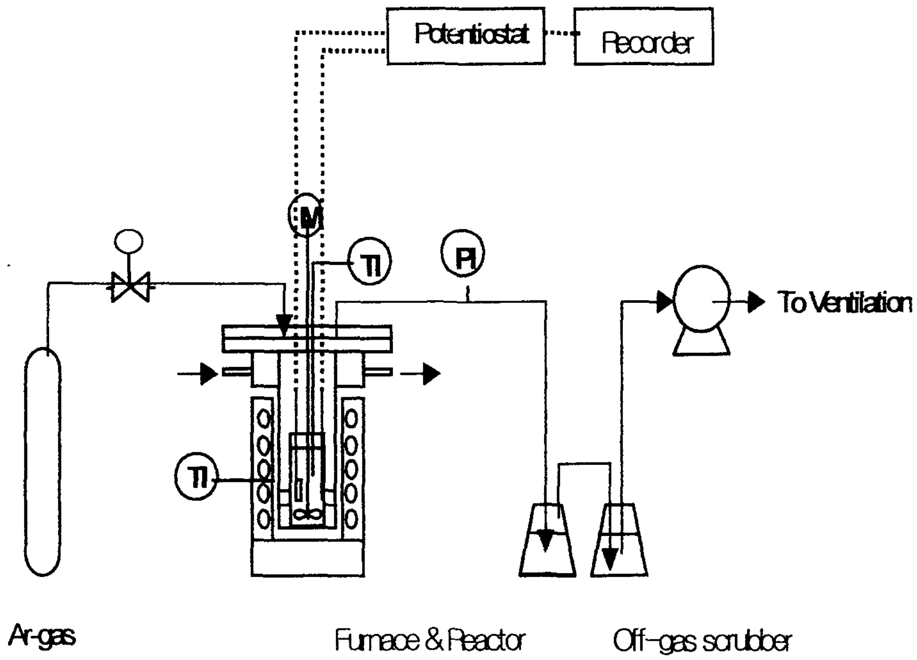


**Figure 2. Schematic diagram of the experimental electrolysis system**

**Figure 3. Deposition of uranium on the carbon steel cathode**

**Electrowinning**

This is also a sort of pyroelectrochemical method to separate some selective metals in a molten salt by electrochemical reduction and deposition on the cathode. One thing different with the electrorefining is that a sacrificing electrode is used as an anode in the electrowinning. Due to the difference in the REDOX potential for the various metal salts just like that in the electrorefining, the anode material must be appropriately chosen for the deposition of desired components. For example, if beryllium is chosen as the anode material in the electrolyte of LiF-$BeF_2$, then uranium, zirconium, and transition metals, having lower change of Gibbs free energy of fluoride formation, tend to deposit on the cathode.

Fig. 4 shows the decomposition potentials of Nb, Zr, U, and Ce in the system of LiF-$BeF_2$(molten salt), Be-anode, and nickel cathode at 500 °C.

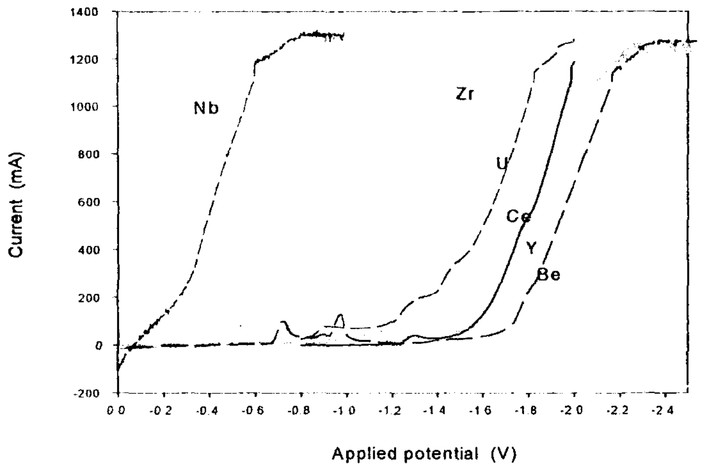


**Figure 4. Decomposition potentials of various metal fluorides in the LiF-BeF2**

## RECOVERY OF TRANSURANIC ELEMENTS

As described above, tansuranic elements are supposed to be recovered as a mixture in the liquid cathode. This would be possible only after removing uranium from the molten salt in advance by the first electrolysis. Since no single plutonium product is obtained in this step, and the mixture of all transuranic elements is to go to the TRU fuel fabrication process, this electro-process could be a part of the proliferation-resistive P-T cycle.

In this step, parts of rare earth tends to accompany transuranic elements into the liquid metal cathode. Therefore, the rare earths should be removed to a certain level that will allow for a transuranic fuel in the transmutation system. Since both rare earth and transuranic elements are distributed into the molten salt and liquid metal phases with different extents, the rare earths can be removed into the salt phase by selective oxidation. At present, a feasibility study is being carried out for the removal of rare earth impurities from the molten cadmium by selective oxidation of rare earths on the basis of thermodynamic properties.

## A STUDY OF TRANSMUTATION SYSTEM

KAERI has been working on ADS since 1997. The KAERI ADS system is called HYPER (HYbrid Power Extraction Reactor) and the conceptual design of HYPER will be completed by 2006. The ADS research of KAERI consists of 3 stages. A basic concept of HYPER was established in the first stage (1997 - 2000) of the development. The basic technology related to HYPER will be investigated in the second stage (2001 - 2003) while upgrading the design. Finally, the conceptual design will be completed in the third stage (2004 - 2006). The investigation of key technologies will be continued in the third stage.

The strategy of experimental study is to join the international collaboration if possible. It is more efficient to get the experimental data and technologies through international collaborations since ADS research is in the early stage in the world. If we need the data and technologies which can not be obtained through international collaborations, then we will do our own experiments. Now KAERI is focused on two main experimental studies, which are Pb-Bi and fuel studies. KAERI joined MEGAPIE project in 2001 and is constructing Pb-Bi corrosion loop. For the fuel study, KAERI is discussing the possible collaboration with ANL-W and investigating fission product irradiation test using KAERI's research reactor HANARO.

### Design Study

HYPER is designed to transmute TRU and some fission products such as I-129 and Tc-99. HYPER is a 1000 $MW_{th}$ system and its $k_{eff}$ is 0.98 [5]. The core consists of 186 fuel assemblies and 24 fission product assemblies. The metal fuel is adopted. I-129 and Tc-99 are put together in the same fission product rod. The TRU fraction of fuel varies according to the core zone. The average TRU weight fractions are 33.6, 40.7 and 44.3% for the inner, middle and outer core respectively. The fuel assemblies are ductless and 13 tie rods exist in one fuel assembly to support fuel rods in the assembly. The tie rod is made of $B_4C$ which also plays a role of a burnable absorber to reduce the reactivity swing [6].

There are 6 safety assemblies to reduce the reactivity swing further. The required current is 10.6 mA at BOC and 17.7 mA at EOC. The inventory of TRU is 5,553 kg at BOC and 302 kg of TRU is transmuted per year. In the case of fission products, I-129 and Tc-99 are transmuted with the rates of 6 and 26 kg/yr respectively. The fuel cycle is 180 days. HYPER adopts scattered fuel reloading system.

MC-CARD, REBUS-3 and DIF3D are used for the core analysis. LAHET code system is used for the target neutronic calculations. KAERI also developed a kinetics code called DESINUR (Design Evaluation and Simulation of Nuclear Reactor). The steady state performance computer codes are developed. MACSIS-H is the code for the analysis of metal alloy fuel and DIMAC is for the dispersion fuel.

Pb-Bi is used as the coolant and spallation target material. The coolant is not separated from the target. The inlet temperature of coolant is 340°C and the average outlet temperature is 490°C. The target window material is 9Cr steel such as T91 and 9Cr-2WVTa. The window has a hemispherical shape with a thickness of 2.5 mm and a diameter of 45 cm. The target system has some requirements for the safe operation. The maximum allowable temperatures of Pb-Bi and beam window are 500°C and 600°C respectively. The maximum allowable stress of the beam window is 160 MPa.

MATRA and SLTHEN are used for the core thermal-hydraulic calculations. CFX and FLUENT are used for the thermal-hydraulic calculations of the target. ANSYS is used for the stress calculation of the beam window.

**Table 1. Basic features of HYPER**

| | |
|---|---|
| Capacity | 1000 $MW_{th}$ |
| $k_{eff}$ | 0.98 |
| Proton Beam | 1 GeV, 10.6/17.7 mA (BOC/EOC) |
| Fuel Type | Metal |
| Coolant/Target | Pb-Bi (Not separated target) |
| Transmutation | TRU, FP (Tc-99, I-129) |
| TRU Tran. Rate | 302 kg/yr |
| FP Tran. Rate | 26 kg/yr (Tc-99), 6 kg/yr (I-129) |
| Support Ratio | 3.3 |

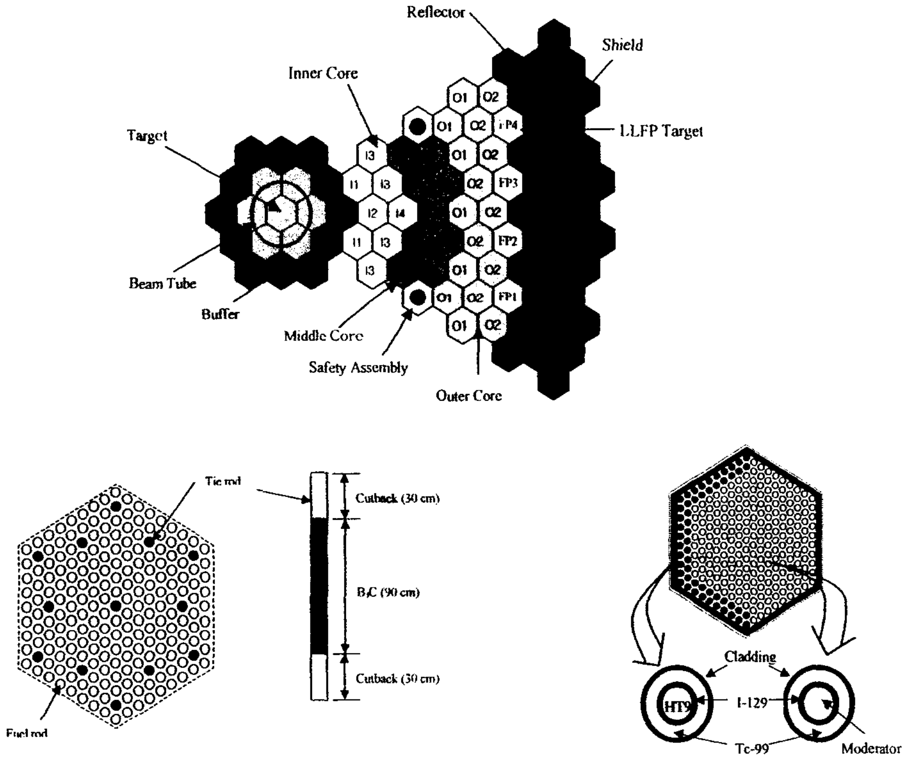


**Figure 5. Assembly design of HYPER**

**Experimental Study**

In addition to the design work, experimental study is being progressed to investigate key technologies. The KAERI's strategy of experimental study is to join international collaboration first for the verification of technologies and possession of the needed data. If we cannot obtain the needed technology and data through the international collaboration, we perform experimental work by ourselves. KAERI is mainly focused on two areas for the experimental study, which are Pb-Bi and fuel development.

KAERI used U to make metal alloy fuel, U-Zr and dispersion fuel, (U-Zr)-Zr. Thermal properties of those U metal fuel was measured and used for studying characteristics of TRU metal fuel. We collaborate with ANL-W for the irradiation test of TRU metal fuel. HYPER fission product target includes both Tc-99 and I-129 in the same rod. We will fabricate the fission product rod and test it using KAERI's research reactor HANARO, which is the 30 MW reactor. Fabrication study will be performed in 2004 and the irradiation will start in 2005.

KAERI joined MEGAPIE project in 2001 for the experimental study of Pb-Bi. MEGAPIE is the one megawatt proton beam irradiation test of Pb-Bi target. PSI, CEA, CNRS, FZK, ENEA, SCK.CEN, KAERI, JAERI and LANL are members of the MEGAPIE project. Now Pb-Bi target is in the stage of fabrication. The irradiation will start in 2005 and PIE will begin in 2006.

The most significant problem in handling Pb-Bi is corrosion. Therefore, KAERI constructed a static Pb-Bi corrosion test facility and is performing experiments using it in 2003. KAERI also started construction of a corrosion loop in 2003 and will complete it in 2004. The temperature range of the loop is between 350°C and 650°C. The oxygen control method is considered to test the protection of steel structure materials against Pb-Bi corrosion.

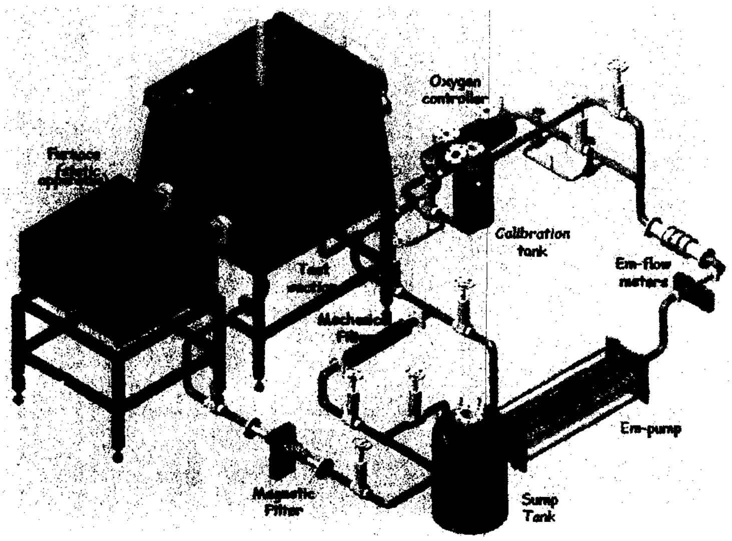


**Figure 6. The schematic drawing of KAERI's Pb-Bi corrosion loop**

## SUMMARY

The R&D activities for partitioning and transmutation of long-lived radionuclides are introduced in this work. An accelerator driven system, named HYPER, is being studied as a candidate transmutation system for the treatment of nuclear wastes. Trans-uranic elements and some long-lived fission products from LWR spent fuel will be the target materials for transmutation. Some part of the conceptual study was already performed in Phase I. The study of Phase II is focused on the evaluation of key unit systems of the HYPER. A conceptual design of the HYPER system will be completed at the end of Phase III. Further studies for the development of pyroprocessing technology will be extended to the employment of transuranic elements in the electrorefining or electrowinning on the basis of international collaboration. As for the HYPER system, the basic core design will be completed in Phase II. More detailed analysis for steady state and transient neutronic behaviors as well as thermal hydraulics in the core will be carried out for various cases including some accidental in Phase III. In-pile tests of simulated fuels will be carried out as an international joint study in the Phase III. HANARO or other research reactors are also considered for the irradiation facility. For the development of cooling system, an experimental loop of Pb or Pb-Bi will be designed and installed in KAERI in order to test corrosive properties of the coolant.

## ACKNOWLEDGMENTS

This study was carried out with the financial support of the Korea National Mid- and Long-Term Nuclear R&D Program by Ministry of Science and Technology.

## REFERENCES

[1] E.J. Karrell, K.V. Gourishankar, L.S. Chow, and R.E. Everhart, "Electrometallurgical Treatment of Oxide Spent Fuels", Global '99, Aug. 29-Sep. 3, Wyoming (1999).

[2] J.J. Laidler, J.E. Battles, W.E. Miller, J.P. Ackerman, and E.L. Carls, Progress in Nuclear Energy, 31(1/2), 131-140 (1997).

[3] T. Inoue, M. Sakata, H. Miyashiro, T. Masumura, A. Sasahara, and N. Yoshiki, Nuclear Technology, 93(Feb.), 206-220 (1991).

[4] J.J. Laidler, J.E. Battles, W.E. Miller, and E.C. Gay, "Development of IFR Pyroprocessing Technology", Global '93, Sep. 12-17, Washington (1993).

[5] Won S. Park, et al., "TRU Incineration Characteristics of Thermal and Fast Subcritical Reactors," Vol. 26, Annals of Nuclear Energy (1999).

[6] Y. H. Kim and W. S. Park, "A Study on Burnable Absorber for a Fast Subcritical Reactor HYPER," 6$^{th}$ OECD/NEA P&T Meeting, Madrid, Spain (2000).