R&D ACTIVITIES FOR PARTITIONING AND TRANSMUTATION IN KOREA

Eung-Ho Kim, Tae-Yung Song, Jae-Hyung Yoo and Hyun-Soo Park
Korea Atomic Energy Research Institute

Abstract

According to the Korean long-term plan for nuclear technology development, KAERI is conducting a R&D project on the partitioning and transmutation (P&T) of long-lived radionuclides. The study for the partitioning is focused on the development of pyroprocessing based on an electrorefining of actinides because it is a kind of proliferation-resistive technology, where all the transuranic metals are separated together as a mixture. The major experimental items of this study include an electrorefining, electrowinning, cadmium evaporation and a molten salt waste treatment. Various behaviours of the electrodeposition of uranium, rare earths, alkali and alkaline earths in the LiCl-KCl electrorefining system have been examined through fundamental experimental work. As liquid cadmium was employed as the second cathode in this study, its removal by an evaporation, thus leaving transuranic as the final product, was examined. Treatment of the molten salt waste was also investigated by introducing zeolite for the immobilization of the molten salts. 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 the 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 the height/diameter, and by introducing the concepts of an optimum core configuration with a transuranic enrichment as well as a scattered reloading of the fuel assemblies.

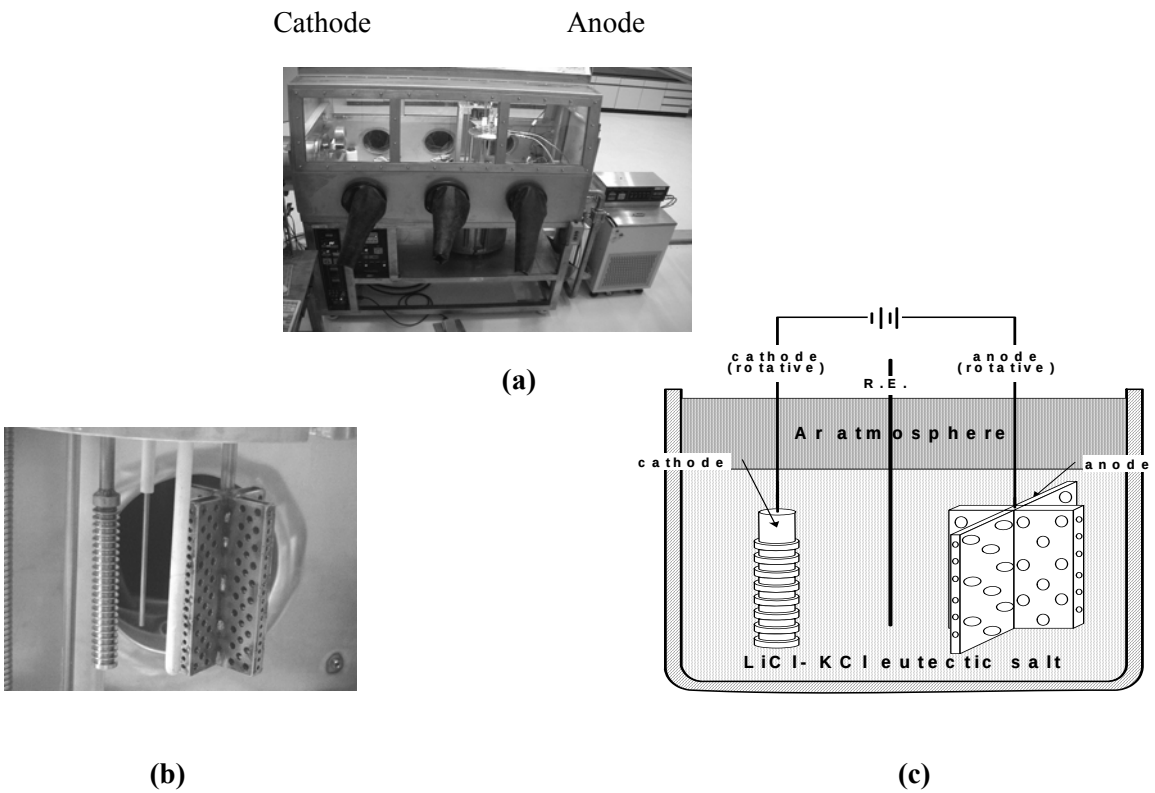
Partitioning of actinide by pyroprocessing

Removal of uranium by electrorefining

Since uranium is not desirable as a fuel source in the transmutation system, it is necessary to remove it from the spent fuels or high-level wastes in advance before recovering the transuranic elements to be transmuted. Electrorefining is a candidate technology to remove the uranium and then to recover the transuranic elements effectively. Since each metal chloride has a unique value of the Gibbs free energy of formation in the molten salt electrolysis, uranium can be selectively reduced and deposited onto the surface of the cathode by the adjustment of the electrical potential between the anode and cathode.

Figure 1 shows the experimental setup of the recent electrorefining system of KAERI. Electrolysis cell consisted of the anode basket containing the metallic fuel, screw type solid cathode and the reference electrode. 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 off-gas from the electrolysis cell is washed with an alkali solution and then water so that the chlorine and hydrochloride, that may be formed in the electrolysis, can be absorbed into the liquids. Some evaporated compounds from the molten salt can also be trapped in the liquids.

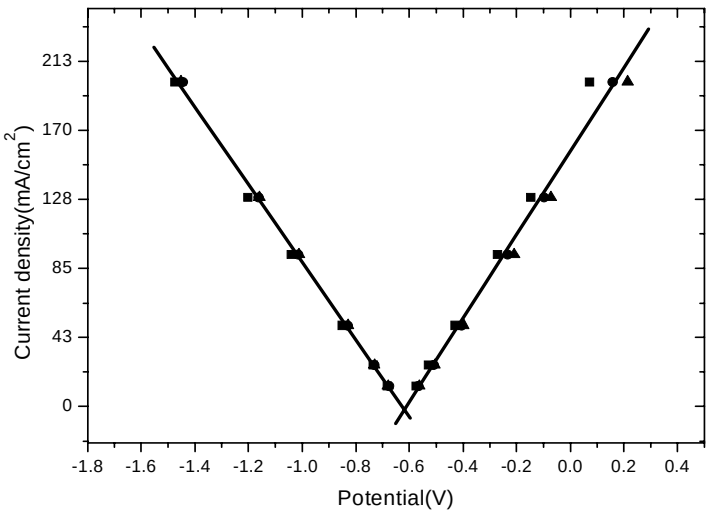
Figure 1. **The experimental setup for the molten salt electrolysis**



(a) glove box system equipped with electrorefiner
(b) anode basket and screw type cathode
(c) electrode configuration

In this work, such operation variables as the rotation speeds of the anode and cathode, structure of the electrode, the initial concentration of uranium in the molten salt, deposited uranium morphology, current density, etc., affecting the electrodeposition of uranium were examined. Among the various experimental work, a typical polarization curve for uranium was obtained as shown in Figure 2. It shows the relationship between the current density and the electrode potentials during the electrolysis. In general, alloy metals containing transition elements are used as the anode basket of the electrorefiner. However, it has been known that corrosion in the anode basket takes place when the potential is greater than 0.4 V for those materials. Accordingly, in order to prevent the anode basket from corrosion during the electrorefining, the current density should be kept at less than 220 mA/cm^2. A typical uranium deposition on the solid cathode at 100 mA/cm^2 shows a well developed dendrite structure.

Figure 2. **Polarisation curves for uranium at the anode and the cathode**



Recovery of the transuranic elements

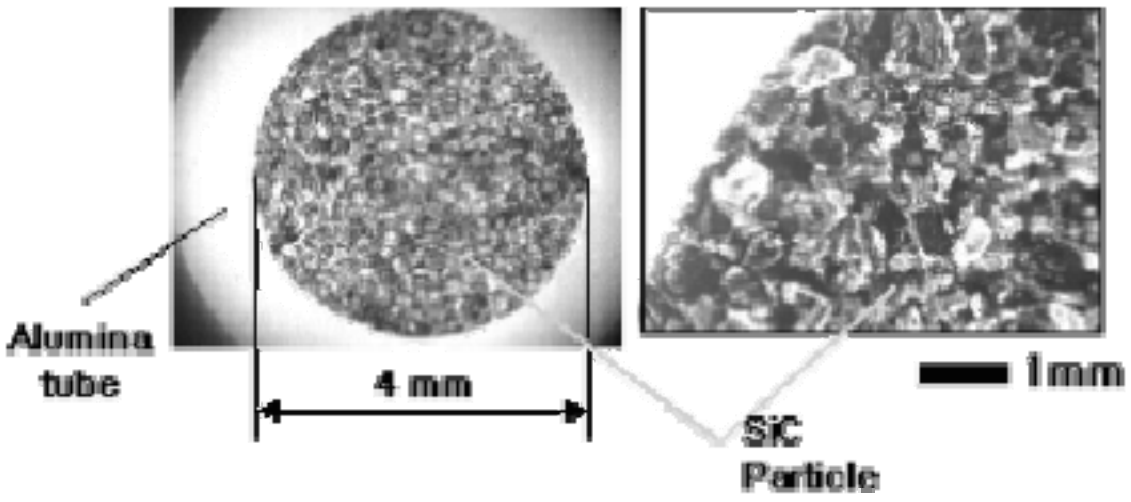
In this work, such rare earth elements as Nd, Ce, and La were used as a surrogate for TRU because they are very close to the TRU elements in the value of the Gibbs free energy of formation in the LiCl-KCl molten salt system. As a result, most amount of the rare earth elements contained in the molten salt (2.8 wt%) were deposited into the moderately agitated liquid cadmium which was used as another cathode. However, most of the deposits were formed at the interface between the salt and cadmium. It seems to be due to the fact that the rare earth elements have a low solubility in liquid cadmium and their density is smaller than that of cadmium but larger than that of the LiCl-KCl salt.

Further studies are also being carried out for the selective oxidation of the rare earth elements that exist in the cadmium phase and then the extraction of them into the molten salt phase in order to find its applicability for the removal of the rare earth elements from the TRU co-deposited into the cadmium.

Preparation of the reference electrode

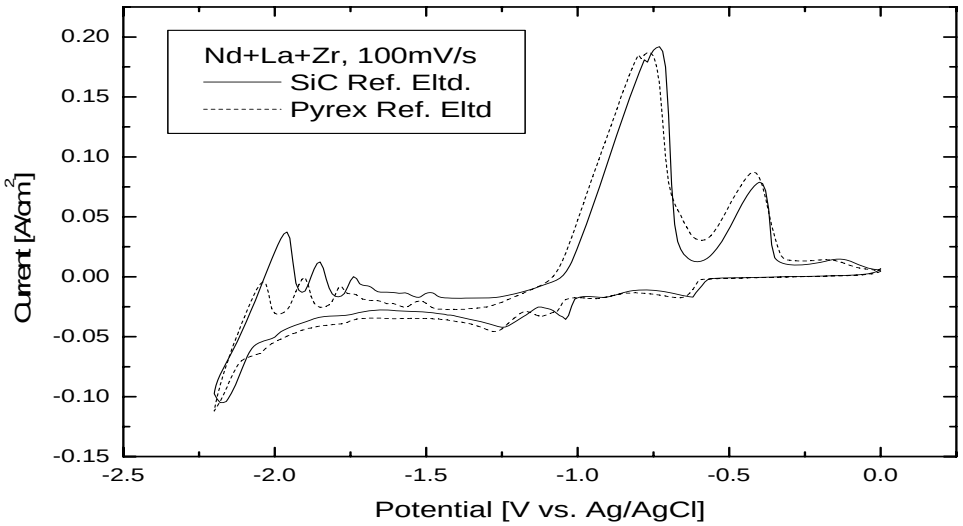
Since the existing reference electrode of Pyrex glass containing silver/silver chloride is quite fragile for handling during the experiments, we tried to develop a solider electrode for a more stable handling. In this work, we replaced the glass tube with an alumina tube and the glass membrane with a silicone carbide (SiC) membrane and then filled the tube with LiCl-KCl eutectic salt containing 1.0 wt% silver chloride where a silver wire was immersed and fixed as a tip of the electrode. Figure 3 shows the cross section of the SiC membrane which was prepared in our laboratory. The powders of SiC and a sintering binder were mixed together, placed at one end of the alumina tube and then sintered at 1 200 for 12 hours so that a porous membrane was formed and clogged the end of the tube. The pore size of this membrane was measured to be about 30 micrometers by means of a mercury porosimeter.

Figure 3. **The reference electrode made of alumina tube and SIC membrane**



The result of the cyclic voltammogram for the new electrode was compared with that obtained for the glass electrode in Figure 4, showing a consistant result in both cases.

Figure 4. **Cyclic voltamograms for the alumina/SiC and Pyrex type reference electrodes**



Design and analysis of the HYPER

Core

HYPER is designed to transmute TRU and some fission products such as 129I and 99Tc. HYPER is a 1 000 MW_{th} system and its k_{eff} is 0.98. Figure 5 shows a schematic configuration of the HYPER core with 186 ductless hexagonal fuel assemblies. As shown in Figure 6, the fuel blanket is divided into 3 TRU enrichment zones to flatten the radial power distribution. In HYPER, a beam of 1 GeV protons is delivered to the central region of the core to generate the spallation neutrons. To simplify the core design, the LBE coolant is used as a spallation target as well. In addition to the ultimate shutdown system (USS), six safety assemblies are placed in the HYPER core for an emergency case. The safety rods are also used conditionally to control the reactivity of the core. For a balanced transmutation of both TRUs and LLFPs (99Tc and 129I), 99Tc and 129I are incinerated in the moderated LLFP assemblies loaded in the reflector zone.

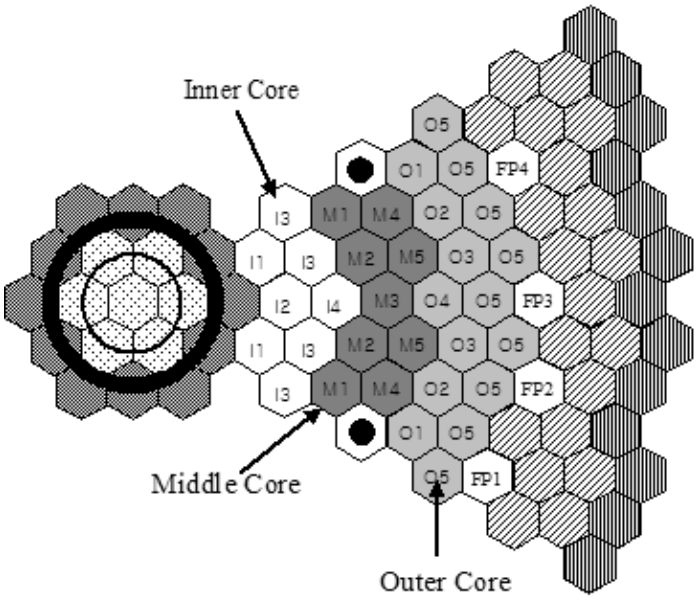
It is well-known that the LBE coolant speed is limited (usually < 2 m/sec) due to its erosive and corrosive behaviour. Therefore, the lattice structure of the fuel rods should be fairly sparse. In fast reactors, a pancake-type core has been typically preferred mainly to reduce the coolant pressure drop. Unfortunately, it has been found that the multiplication of the external source is quite inefficient in a pancake type ADS because of the relatively large source neutron leakage. It was shown that the maximum source multiplication can be achieved when the core height is about 2 m [1]. Taking into account the source multiplication and the coolant speed, the core height of HYPER was compromised at 150 cm, and the power density was determined such that the average coolant speed could be about 1.65 m/sec. To reduce the core size and improve the neutron economy, a ductless fuel assembly is adopted in the HYPER system. An advantage of the ductless fuel assembly is that the flow blockage of a subassembly is basically impossible and the production of activation products in the duct can be avoided.

Concerning a TRU-loaded ADS using a fixed cycle length, one of the challenging problems is a very large reactivity swing, leading to a large change of the accelerator power over a depletion period. Even in an ADS loaded with a MA (Minor Actinide) fuel, the burnup reactivity swing is found to be fairly noticeable, although it is relatively smaller than that in a TRU-loaded core. The large burnup reactivity swing results in several unfavorable safety features as well as deleterious impacts on the economics of the system. In the HYPER core, the B-10 was also used as a burnable absorber (BA) in a unique way to reduce the reactivity swing and control the core power distribution [2].

The required current is 10.6 mA at BOC and 16.4 mA at EOC. The inventory of TRU is 6 510 kg at BOC and 282 kg of TRU is transmuted per year. In the case of fission products, 129I and 99Tc are transmuted with the rates of 7 and 27 kg/yr respectively. The fuel cycle is 180 days. HYPER adopts a scattered fuel reloading system.

MC-CARD, REBUS-3 and DIF3D are used for the core analysis. The 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).

Figure 5. **Schematic diagram of the HYPER core**

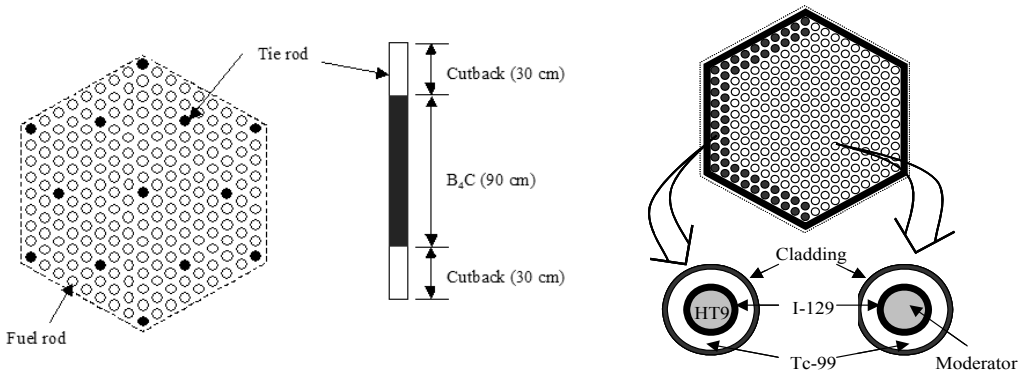


Fuel and Fission Product

In general, a non-uranium alloy fuel is utilized in a TRU transmuter to maximise the TRU consumption rate. Previously, a Zr-based dispersion fuel was used as the HYPER fuel since it was expected that a very high fuel burnup could be achieved. However, we have found that the dispersion fuel transforms to a metallic alloy during a high temperature operation. Therefore, in the current design, a metallic alloy of U-TRU-Zr is utilized as the HYPER fuel, in which pure lead is used as the bonding material. As a result, a large gas plenum is placed above the active core.

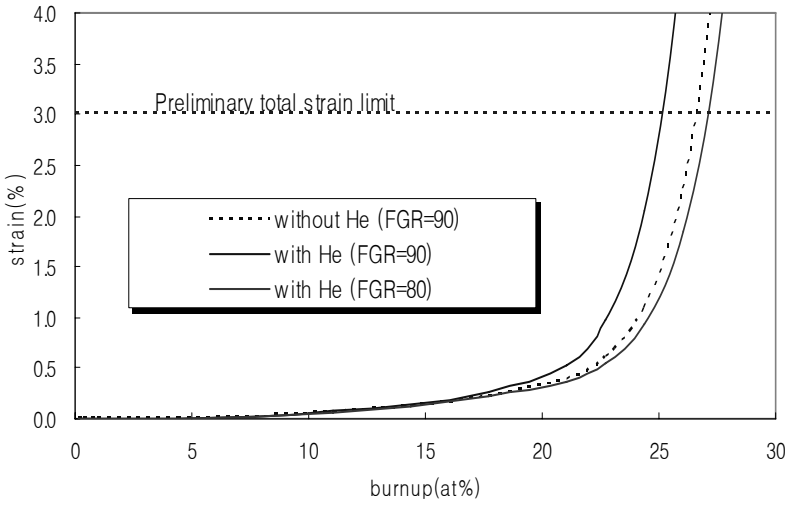
Each fuel assembly has 204 fuel rods and the fuel rods are aligned in a triangular pattern with 13 tie rods. A fairly open lattice with a pitch-to-diameter (P/D) ratio of 1.49 is adopted in HYPER. In Figure 6, a schematic configuration of the ductless fuel assembly is shown. The B-10 burnable absorber is loaded into the tie rods with top and bottom cutbacks in order to enhance the B-10 depletion rate and also to flatten the axial power distribution of the core. The BA concept with the cutbacks can effectively mitigate the peak fast neutron fluence of the assembly. The peak fast neutron fluence is a limiting design criterion in the LBE-cooled fast reactors.

Figure 6. **Fuel and fission product assemblies**



The MACSIS-H for an alloy fuel and the DIMAC for a disperison fuel are being developed as the steady-state performance analysis code, respectively. Main structures of each code consist of the temperature profile calculation routine, the swelling/FGR calculation routine, and the deformation calculation routine. The He production rates calculated by the other code are inserted into the swelling/FGR routine of each code. Figure 7 is one example of the MACSIS-H calculation. The strain was calculated as a function of the burnup of the HYPER fuel. The calculation was performed with and without a He generation by varying the fission gas rate. The result shows that the maximum strain is lower than the limit of the HYPER average discharge burnup of 17 at %.

Figure 7. Strain vs burnup for the HYPER fuel

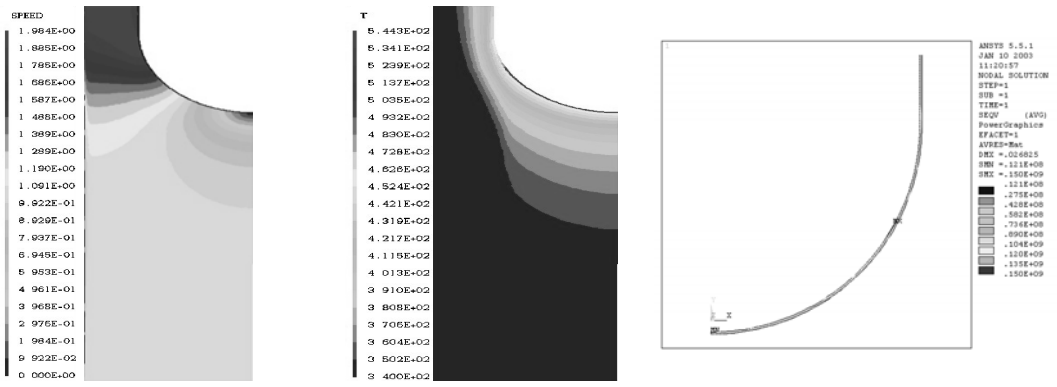


Coolant and Target

Pb-Bi is used as the coolant and spallation target material. The coolant is not separated from the target. MATRA and SLTHEN are used for the core thermal-hydraulic calculations. SLTHEN can be used for a multi-assembly analysis. Therefore, a 7 and 45 assembly analysis of HYPER was performed using SLTHEN. MATRA was developed to be used for the ductless assembly with a grid spacer, which is the case of the HYPER fuel assembly. The sub-channel analysis was performed using MATRA and the result shows that the average outlet temperature is 490^{o}C when the inlet temperature of the coolant is 340^{o}C. The maximum cladding temperature turned out to be 570^{o}C.

The cylindrical beam tube and the hemispherical beam window were adopted in the basic target design concept with a 1 GeV proton energy, and the thermal hydraulic and structural analyses were performed with the CFX and ANSYS codes. The target window material is 9Cr steel such as T91 and 9Cr-2WVTa. The beam window diameter and thickness were varied to find the optimal parameter set based on the design criteria: maximum Lead-bismuth eutectic (LBE) temperature < 500^{o}C, maximum beam window temperature < 600^{o}C, maximum LBE velocity < 2 m/s, and the maximum beam window stress < 160 MPa. The results show that a 40 cm wide proton beam with a uniform beam profile should be adopted for HYPER. It was found that a 2.5 mm thick beam window is needed to sustain the mechanical load. When the inlet velocity of Pb-Bi is 0.95 m/s, the maximum allowable current is 24.1 mA, which is greater than the required current of HYPER.

Figure 8. **CFX and ANSYS results of the HYPER target calculation**



Velocity (Max=1.96 m/s) | Temperature (Max=544 ^{o}C) | Stress (Max=150 MPa)

Experimental

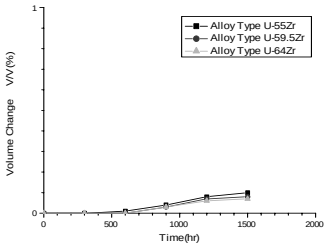
Fuel

Two types of metal fuels were considered for the HYPER fuel. One is a metal alloy type and the other is a dispersion type. Both types of U surrogate fuel samples were fabricated and the basic characteristics were investigated. It was found that the dispersion fuel was not made with a good microstructure and the original structure of the dispersion fuel was not kept after an annealing. Therefore, we chose the metal alloy fuel for the HYPER fuel type.

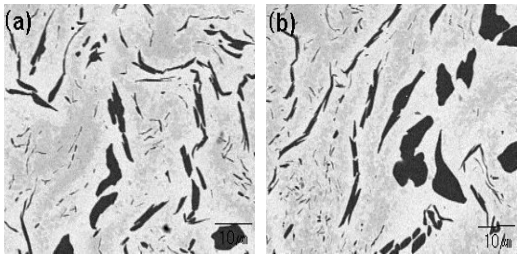
The reference blanket fuel pin of HYPER consists of the fuel slug of the TRU-xZr (x=50-60wt%) alloy and it is immersed in lead for a thermal bonding with the cladding. The blanket fuel cladding material is ferritic-martensitic steel HT-9. As a basic study on the HYPER fuel, we fabricated surrogate U-50, 55, 60wt%Zr alloy fuel instead of the actual TRU-Zr fuel. The U-Zr metallic fuel was fabricated by mixing, pressing, sintering and extrusion. The sintering temperature and time were 1500°C and 2 hrs respectively.

After fabricating the surrogate fuel, the thermal properties such as the thermal conductivity and the thermal expansion coefficient were measured. We also performed a thermal stability test of the surrogate fuel to investigate the volume change and the microstructure change. The fuel samples were put into the furnace for 1 500 hrs with the temperature of 630°C and 700°C. Figure 9 shows the volume change as a function of the time and the microstructure change after an annealing. Reaction characteristics were also investigated among the Pb bonding, cladding material (HT-9) and U surrogate fuel.

Figure 9. **Volume and microstructure change of the U-Zr sample after an annealing**



Volume change



Microscopic structure : (a) Before (b) After

Pb-Bi

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

The most significant problem in handling Pb-Bi is corrosion. Therefore we performed static corrosion tests using FZK's facility COSTA to investigate the dissolution effect. KAERI also installed a static corrosion facility in 2003 and started static corrosion tests. Figure 10 shows the schematic diagram of the static corrosion facility. It is mainly composed of tube furnaces, a gas system and a glove box. The furnace has three independent zone heaters to reduce the temperature difference.

The test materials were 316LN and some ferrite/martensitic steels such as HT-9 and T91. The test was performed under both reduced and oxygen-controlled atmospheres. Part of the test results are shown in Figure 10.

Figure 10. **Schematic diagram of the KAERI static corrosion facility and test results**

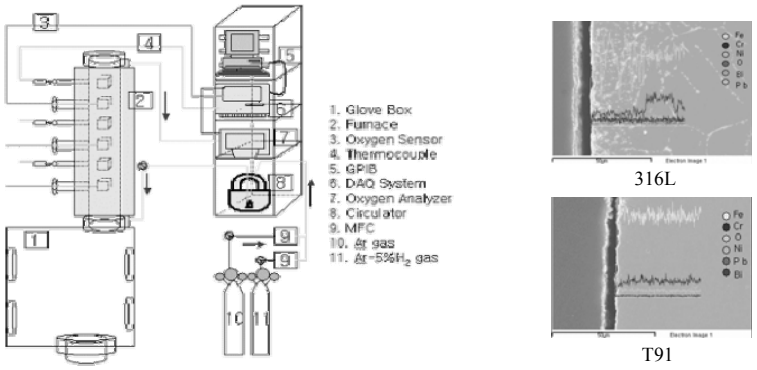
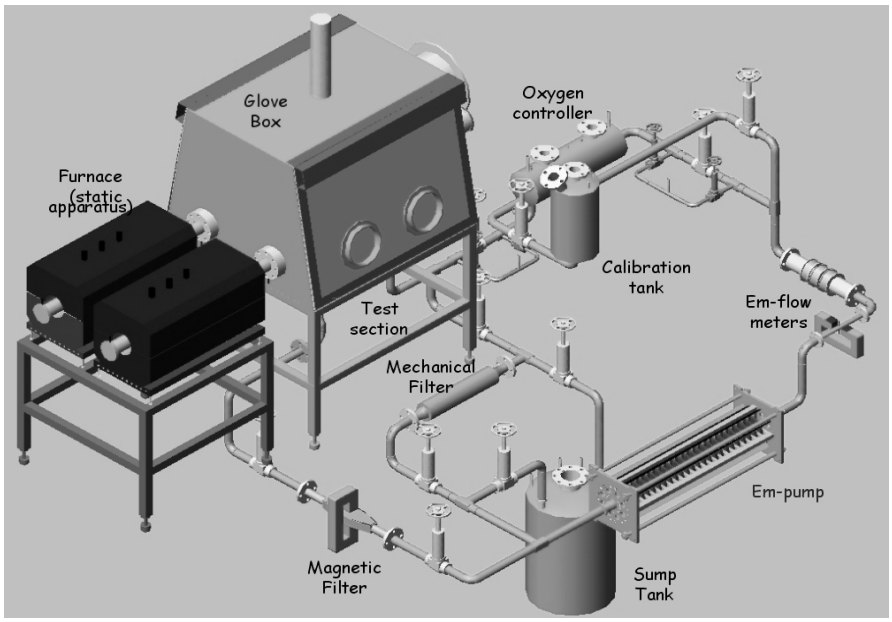


Figure 11 shows the schematic diagram of the dynamic corrosion loop to be installed at KAERI. The LBE loop is an isothermal loop. The flow velocity in the test section was designed to be around 2m/s in the range of 400 ~ 550°C and the charging volume of the LBE is around 0.03 m^3 in the circulation loop.

Figure 11. **Three dimensional schematic diagram of the corrosion loop**



The LBE loop is mainly composed of a main test loop, bypass loop for filtering the LBE and a mixture gas supplying system. The liquid metal in the main test loop circulates in the following order: EM pump – EM flow meter – oxygen controller – test section – magnetic filter – EM pump. From the analysis of the pressure drop, the specification of the piping system was determined as a 1.5 inch pipe to reduce the pressure drop by a high mean fluid velocity. The pressure drop of the main test loop was estimated at around 3 bar with the flow rate of 60 lpm.

The oxygen concentration in the range of 10^{-7}wt% ~ 10^{-5}wt% is controlled by the chemical equilibrium between the mixture gas of hydrogen-argon and the water vapor. At present, the oxygen concentration in the LBE and the mixture gas is measured with an oxygen sensor made of Yttria Stabilized Zirconia as a solid electrolyte cell and Pt/air as a reference system.

Summary

KAERI has been working on ADS since 1997. The KAERI ADS system is called HYPER (HYbrid Power Extraction Reactor). HYPER research started as a 10 year nuclear research programme. The ADS research of KAERI consists of three stages. The conceptual design of the core was almost completed in the second stage (2000-2003). The core design will be upgraded and modified in the third stage (2004-2006). For example, the dual annular injection tube will be introduced to reduce the flow rate of the Pb-Bi in the target channel. The structure analysis of the HYPER fuel assembly will be also performed. But the main work related to the design and analysis is a transient case study. Some core transient cases such as LOHS-WS and LOF-WS will be studied. The transient cases related to the target, beam window and fuel assembly structure will also be studied. KAERI fabricated an U surrogate metal fuel and performed tests using the U fuel sample to investigate the basic characteristics in the second stage of research. The HYPER fission product target includes both 99Tc and 129I in the same rod. We will fabricate the fission product rod and test it using KAERI's research reactor HANARO, which is a 30 MW reactor. Fabrication study of the FP target will be performed in 2004 and the irradiation will start in 2005. KAERI will complete the construction of a corrosion loop in 2004. The oxygen control method is considered to test the protection of steel structure materials against Pb-Bi corrosion. Therefore, an oxygen sensor will be developed. KAERI will launch the I-NERI programme in June, 2004. The lead-alloy corrosion will be investigated through the I-NERI programme. The period of I-NERI is three years and LANL is the U. S. partner.

Acknowledgements

The Korean Ministry of Science and Technology (MOST) has supported this work.

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[2] Y. Kim *et al., Core Design Characteristics of the HYPER System*, OECD/NEA 7th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Jeju, Korea, 14-16 October 2002.