R&D ACTIVITIES FOR PARTITIONING AND TRANSMUTATION IN KOREA

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Abstract

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 in the areas of partitioning and transmutation system. The R&D activities for partitioning and transmutation of long-lived radionuclides are introduced in this work. The studies of partitioning are focused on the electrorefining and electrowinning, which are aimed at investigating the thermodynamic properties of electrodeposition behaviours as well as the separation efficiency. As for the transmutation system, the HYPER (HYbrid Power Extraction Reactor) combined by a proton accelerator and a sub-critical reactor is being studied in KAERI as a prominent candidate facility in the future. Some conceptual studies are being conducted to develop key elemental systems of the sub-critical reactor such as the core, TRU fuel, proton target, and the cooling system. The conceptual design of the HYPER system will be completed by 2006.

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 (12 PWRs and 4 PHWRs) are currently in operation, supplying about 40% of total electricity demand in Korea. As of December 2001, the accumulated spent fuel accounts for 5 385 t that is stored at four reactor sites. The cumulative amount is prospected to reach 11 000 t 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 and 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 sub-critical 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 sub-critical 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 ^{99}Tc and ^{129}I 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 analysing 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 programmes.

Strategy of partitioning and transmutation

The nuclear transmutation, though it is not fully developed yet for commercialisation, 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 programme, 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.

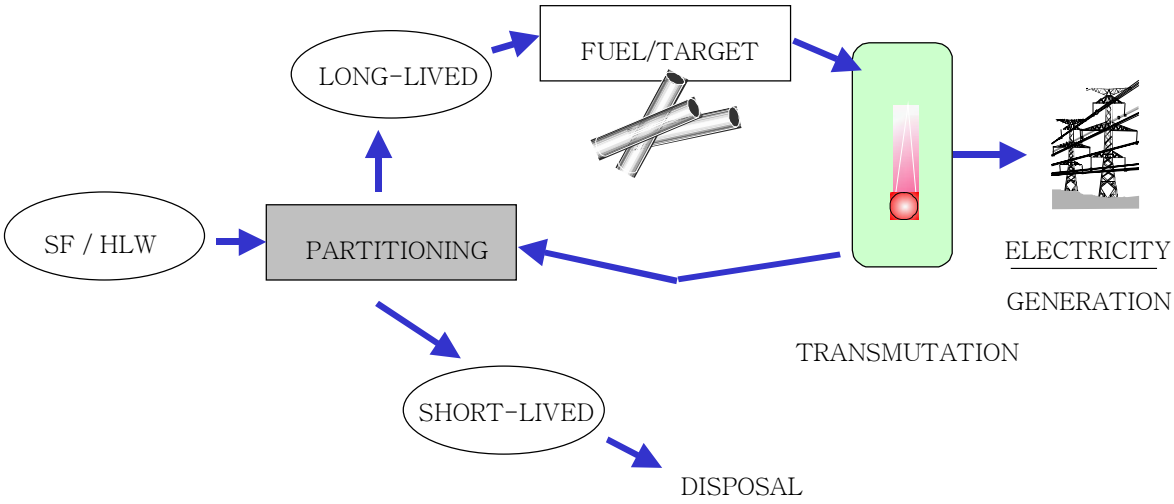
Conversion of oxide fuel material is being taken into account in order to reduce the oxides into metallic forms by using lithium in a lithium chloride bath. The materials of metallic forms then will be

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 ^{129}I and ^{99}Tc 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 Figure 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 summarised as follows:

- Exclusive use of uranium as a fuel source in the transmutation system.
- Partitioning based on non-proliferation.
- 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 effect, 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.

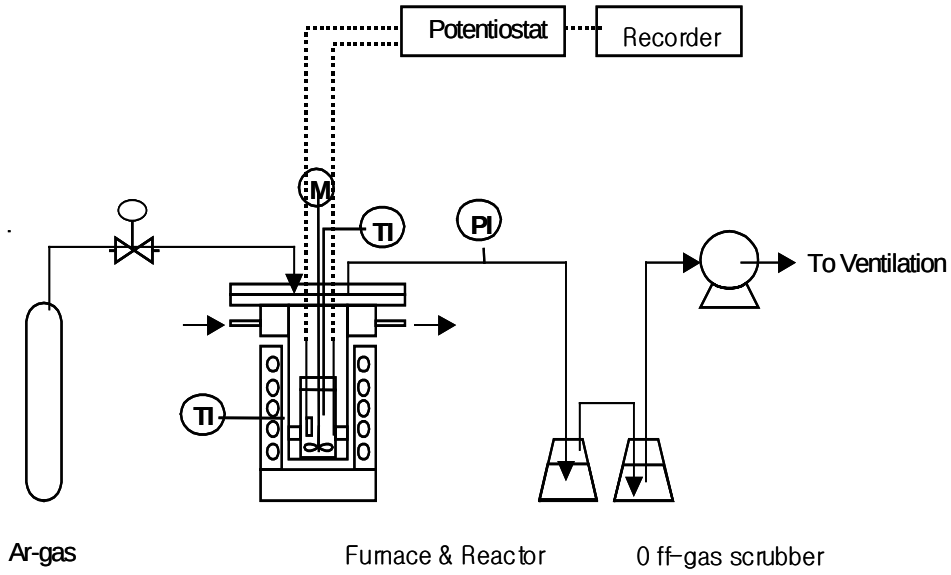
Electrorefining

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.

Figure 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.

Figure 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 experimental electrolysis system**



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.

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

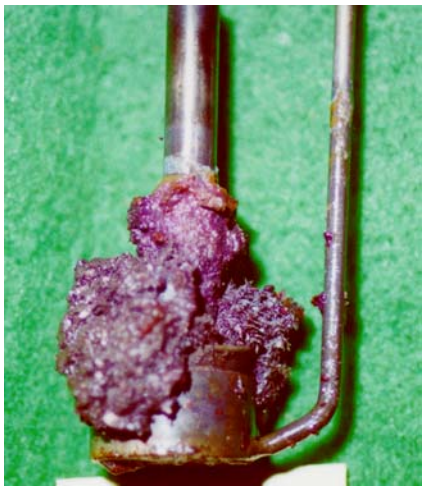
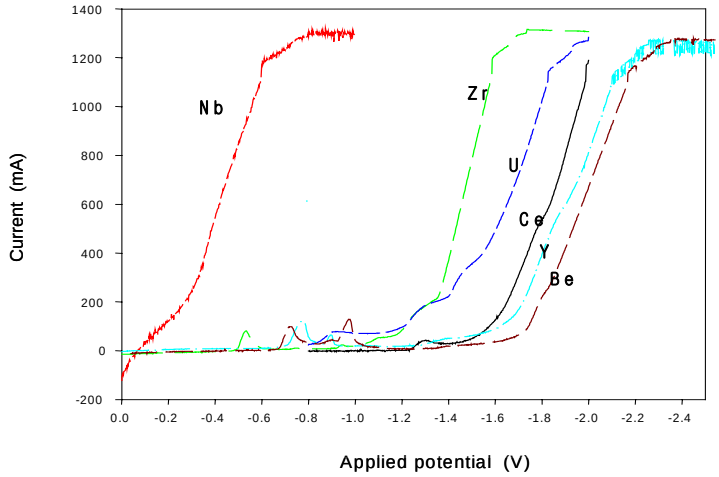


Figure 4 shows the decomposition potentials of Nb, Zr, U, and Ce in the system of LiF-BeF_2(molten-salt), Be-anode, and nickel cathode at 500°C.

Figure 4. **Decomposition potentials of various metal fluorides in the molten-salt of LiF-BeF_2**



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 conceptual study of transmutation system

An accelerator driven system, named HYPTrans-uranics(TRU) and some long lived fission products(FP) from LWR spent fuel will be the target materials for transmutation. The fission products that deserve the most attention are ^{99}Tc, ^{135}Cs, and ^{129}I from the viewpoint of releasing risk from a high-level waste repository. Since cesium separated from spent fuels, unlike ^{99}Tc and ^{129}I, is very hard to be transmuted, ^{99}Tc and ^{129}I are the objective fission products considered as transmutation targets in the HYPER system.

Some part of the conceptual study was already performed in Phase I (1997-2000). The study of Phase II (2001-2003) 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 (2004-2006).

HYPER core

The HYPER is being considered as one of the prominent transmutation systems to treat TRU as well as long-lived fission products at the same time. Basic core design parameters were derived as follows in Phase I:

- Neutron energy spectrum and pitch-to-diameter ratio.
- TRU fuel loading scheme and its optimisation.
- Methodology development to minimise the reactivity swing.

The HYPER will be designed as a fast neutron-irradiation system because fast neutron is much more efficient for the transmutation of TRU in terms of accelerator beam power economy, pin power peaking, and transmutation capability for all transuranics. [5] In order to keep the relative assembly power within the design target value of 1.5, the core should be divided into three different TRU enrichment zones. The fuel of low TRU fraction (26%TRU-74%Zr) will be designed to load in the innermost zone whereas the fuel of high TRU fraction(46%TRU-54%Zr) will be loaded in the outermost region. Refueling will be carried out on the basis of the scattered loading concept with multiple batches in each zone. In this case the use of burnable absorbers in the core was found to reduce the reactivity swing by 38%. [6]

Various optimisation studies on the concept of core are now underway in Phase II as follows:

- Optimisation of height-to-diameter ratio. [7]
- Optimisation of sub-criticality level.
- Optimisation of spallation source.
- Development of 3-D kinetics code, HITE (3-dimensional hexagonal finite element code for transient and steady state).

In the analysis of HYPER system, the partitioning process is assumed to achieve the rates of uranium and lanthanide eliminations from LWR spent fuel (discharged burn-up ≅ 33 000 MWD/MTU) as 99.9% and 90%, respectively. As a result, the ratio of TRU to uranium will be 9 to 1 in the HYPER. With this ratio, the HYPER core will reach a pseudo-equilibrium condition in terms of nuclide composition, approximately at the 20^{th} cycle. The fraction of uranium in heavy metal will be about 20% at the 20^{th} cycle. The transmutation capability of the system has been evaluated as about 313 kg of TRU for one effective full power year. About 90% of energy will be generated from the fission of TRU while the other 10% will be from the uranium at 20^{th} Cycle. Table 1 shows the actinide burning characteristics of the HYPER at the 20^{th} cycle.

Table 1. **Variation of actinide concentration at the 20th cycle**

Nuclide	**Inventory** (kg)		
	BOC	EOC	Variation
^{233}U	0.2144E-02	0.1902E-02	-0.0002
^{234}U	0.9166E+01	0.9215E+01	+0.039
^{235}U	0.3203E+01	0.3121E+01	-0.082
^{236}U	0.3949E+01	0.3986E+01	+0.037
^{238}U	0.9675E+03	0.9516E+03	-15.9
^{237}Np	0.7585E+02	0.6977E+02	-6.08
^{238}Pu	0.1456E+03	0.1432E+03	-2.40
^{239}Pu	0.8774E+03	0.8082E+03	-69.2
^{240}Pu	0.1299E+04	0.1270E+04	-29.0
^{241}Pu	0.2797E+03	0.2674E+03	-12.3
^{242}Pu	0.3898E+03	0.3845E+03	-5.3
^{241}Am	0.1049E+03	0.1010E+03	-3.9
^{242}Am	0.7905E-01	0.7898E-01	-0.0001
^{242m}Am	0.9818E+01	0.9773E+01	-0.045
^{243}Am	0.1191E+03	0.1182E+03	-0.9
^{242}Cm	0.8502E+01	0.8498E+01	-0.004
^{243}Cm	0.1090E+01	0.1081E+01	-0.009
^{244}Cm	0.1142E+03	0.1142E+03	0.0
^{245}Cm	0.3564E+02	0.3571E+02	+0.07
^{246}Cm	0.3084E+02	0.3072E+02	-0.12

The sub-criticality level of the core has been determined by considering the reactivity changes due to the power defect, beam tube rupture. The analysis showed that the sub-criticality of 0.97 is enough to keep the core under criticality at any anticipated condition. The HYPER system has large reactivity swing because it has very little fertile material. The reactivity swing limits the cycle length to be about 140 days. Figure 5 shows a brief description of the overall core design concept. Table 2 represents the design parameters for the HYPER system.

Figure 5. **HYPER core design concept**

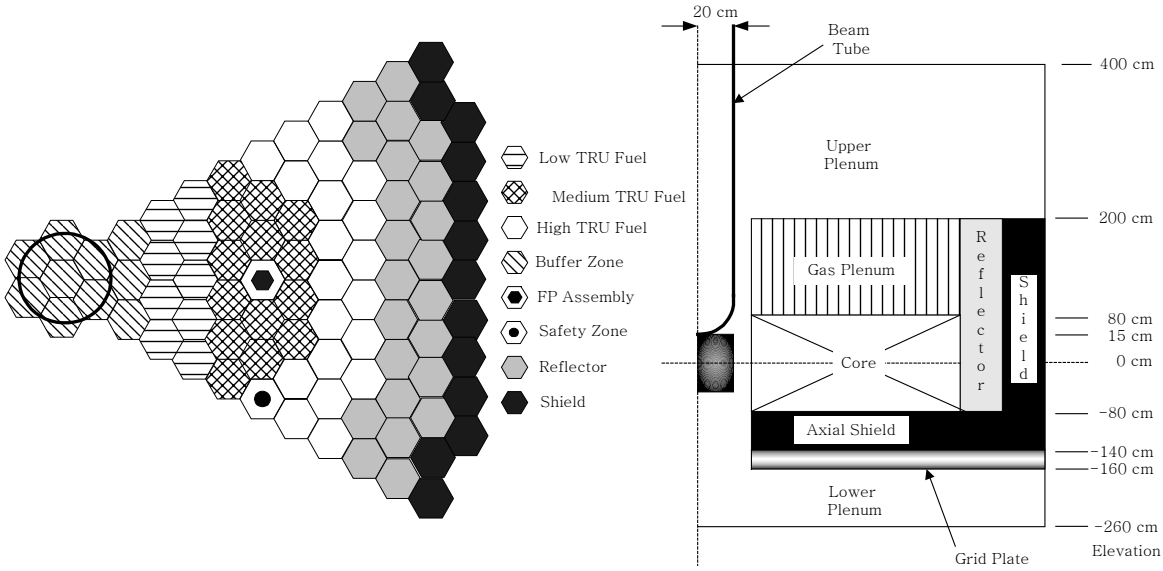


Table 2. **HYPER system design parameters**			
Parameter (unit)	**Values**	**Parameter** (unit)	**Values**
System		***Assembly/Fuel***	
• Core Thermal Power (MW)	1 000	• Ass. Pitch (cm)	19.96
• Active Core Height (m)	1.2	• Flow Tube Outer Surface Flat-to-Flat Distance (cm)	19.52
• Effective Core Diameter (m)	3.8		0.355
• Total Fuel Mass (TRU-Kg)	2 961	• Tube Thickness (cm)	HT-9
• System Multiplication Factor	0.97	• Tube Material	331
• Accelerator Beam Power (MW)	~ 13	• Rods per Assembly	1.5
• Ave. Discharge Burnup (%at)	~ 25	• Rod P/D	0.668
• Transmutation Capability (Kg/EFPY)	313	• Rod Diameter (cm)	0.492
	183	• Fuel Meat Diameter (cm)	0.051
• Number of Fuel Assembly	13.5	• Cladding Thickness (cm)	
• Ave.Linear Power Density (KW/m)			

HYPER fuel

One of the basic requirements for the selection of HYPER fuel is a good compatibility with the partitioning process. As the loss of radioactive nuclides cannot be avoided at each recycling process, a high burn-up capability is required as a selection criterion in order to minimise the loss. A metallic fuel, among other fuel types such as oxides and nitrides, has been selected because it has relatively good comptabilities with the dry partitioning process, high burn-up requirement, and also fast neutron spectra. Either a TRU-Zr alloy or a (TRU-Zr)-Zr dispersion-type is considered as a candidate fuel for the HYPER system. In the case of the dispersion fuel, particles of 90wt%TRU-10wt%Zr metal alloy are to be dispersed in a Zr matrix.

Two computer simulation codes, MACSIS-H (alloy-type fuel analysis) and DIMAC (dispersion-type fuel analysis), were developed in Phase I. In addition, a basic fuel design and a couple of fuel performance analyses have been done using the codes. [8,9] Since there is not much knowledge on TRU-Zr type metal fuel most of the experimental result was reflected in the modification of the codes. The increment of Zr fraction decreases the deformation rate of a fuel rod (ΔD/D; D=Fuel Rod Diameter) in the alloy-type fuel as expected. The simulation predicted that more than 90% of the fission gas is released at burn-up of 5 atomic per cent for the alloy-type. The fission gas release rate is almost independent of the Zr fraction when Zr weight fraction varies from 45% to 55%. The build-up pressures due to the fission gas are 818psig and 553psig when the plenum volumes are 1.2 and 1.5 times the fuel meat volume, respectively at the burn-up of 13.6 atomic percent. Both results are acceptable. In addition to the fission gas, helium gas is produced in the TRU fuel because of the alpha decay of ^{241}Am, ^{242}Cm. Thus, larger plenum size (1.5 times) is preferred in the metal fuel bearing TRU.

Most of the fission gas is modelled to be trapped at the inside of the fuel matrix for the dispersion-type fuel. Therefore, the dispersion fuel does not have gas plenum. The trapping of fission gas in the fuel matrix changes the fuel volume. The simulation predicts that the dispersion-type fuel has the deformation rate (ΔD/D) of 3.5% at 30at% burn-up.

In addition to the continuous update of the computer codes, simulated fuels (U-Zr) and ((U-10Zr)-Zr) have been fabricated and tested under the out-pile condition. Basic properties such as thermal conductivity, expansion, and density were obtained. Thermal stability, the reaction between fuel meat and cladding, and the redistribution of nuclides are under examination. Figure 6 and 7 show the U-10Zr powder fabricated using a centrifugal method and the measured thermal conductivity/expansion coefficients, respectively.

Pb-Bi Coolant and target

A lead-bismuth (Pb-Bi) eutectic alloy was selected as a coolant material for the HYPER system because Pb-Bi is rather safe in terms of chemical reactivity and can be used as a spallation target also. A preliminary design study was performed for its thermal hydraulics in the core in Phase I.

Figure 6. **Particle shape and size distribution of U-10Zr**

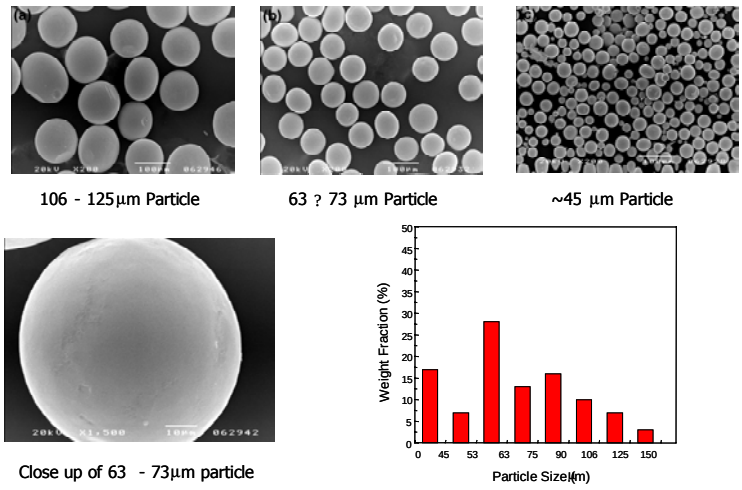
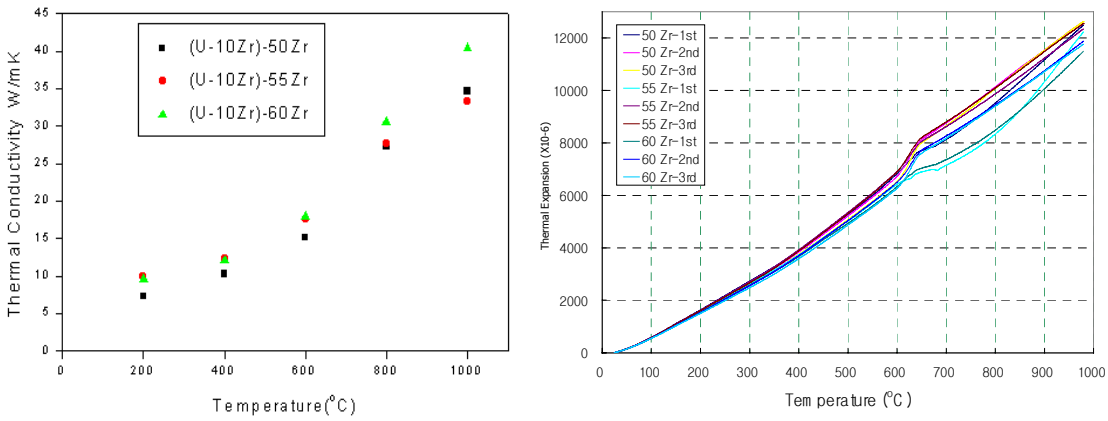


Figure 7. **Thermal conductivity and expansion coeff. of (U-10Zr)-Zr) type fuel**



The core inlet and outlet temperatures of Pb-Bi coolant have been determined as 340°C and 510°C, respectively by considering the melting point of Pb-Bi and the corrosion of the structural material, etc. The P/D (Pitch-to-Diameter) ratio of the HYPER core was chosen as 1.5 and the average Pb-Bi velocity as 1.5 m/s, which is a relatively low coolant velocity compared to that of typical LMR reactors using sodium as coolant. [10] The flowing velocity of Pb-Bi is limited to be less than 2.0m/sec in order to avoid any possible erosion problems and reduce the required pumping power. Instead of wire spacers commonly used for tight lattices, grid spacers are more suitable to ensure proper separation of the fuel rod in the HYPER system. A loop type configuration was adopted and a 3-loop concept was estimated to be the optimum. Each loop has one super-heater and two evaporators. Figure 8 and Table 3 show the design characteristics of cooling system of the HYPER.

Figure 8. **Heat removal system concept for the HYPER system**

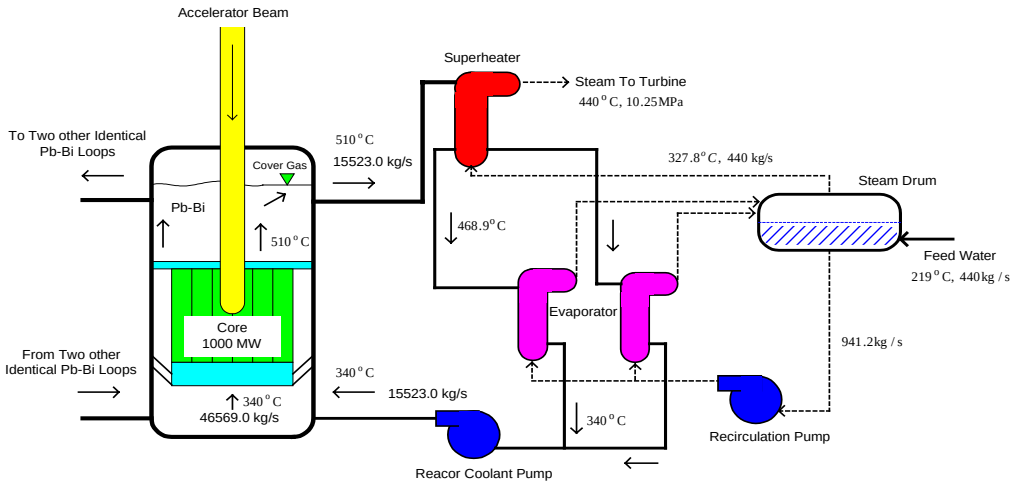


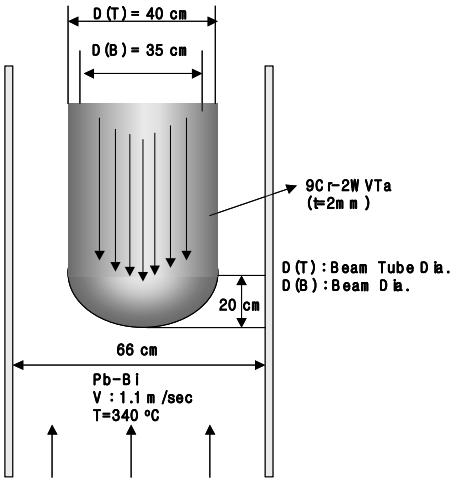
Table 3. **Thermal hydraulic design parameters for the HYPER system**

Thermal Power	1 000 MWt
Primary Heat Transport System	
Reactor Inlet/Outlet Temperature	340°C/510°C
System Flow Rate	40 155 kg/sec
Number of Loop	3 Loop
Primary Coolant Pump	Mechanical Type
Steam Generator	
Type	Separate Type (Evaporator/Superheater)
Superheater	Once-Through Type, Single Wall Tube
Thermal Power	132.8MWt
Primary Side	
Inlet/Outlet Temperature	510°C/465°C
Mass Flow Rate	20 077 kg/sec
Secondary Side	
Operating Pressure	156.7 Atm
Inlet/Outlet Temperature	347°C/490°C
Mass Flow Rate	195.2 kg/sec
Evaporator	Forced Circulation Type, Single Wall Tube
Thermal Power	367.2MWt
Primary Side	
Inlet/Outlet Temperature	465°C/340°C
Mass Flow Rate	20 077 kg/sec
Secondary Side	
Recirculation Ratio	5
Operating Pressure	156.7 Atm
Inlet/Outlet Temperature	312°C/347°C
Feedwater Flow Rate	195.2 kg/sec
Circulation Flow Rate	976 kg/sec

The Pb-Bi coolant flowing through the central channel will be used as the spallation target. The factors affecting the lifetime of the beam window are corrosion by Pb-Bi and radiation damage. The alloy 9Cr-2WVTa is being considered as a window material because the advanced ferritic/martensitic steel is known to have a good performance in a highly corrosive and radiative environment. A single beam window is adopted so that there is no independent window cooling system required. The design target for the lifetime of the window is no less than 6 months. The average energy of the spallation neutrons is estimated to be 14 MeV. The spallation reaction produces more than 800 different radioactive nuclides. One of the dominant long-lived radioactive nuclides due to the spallation is ^{205}Pb. About 9.3 grams of ^{205}Pb is generated when 1 GeV, 1 mA protons are injected into the Pb-Bi target for one effective full power year. Figure 9 shows the lay out of beam target zone.

A basic design analysis is being carried out using SLTHEN (Steady-state LMR core Thermal Hydraulics code based on ENergy model) [11] for core sub-channel analysis and CFX for the analysis of the thermal hydraulics in the target region. The maximum coolant outlet temperatures in the case of the hotest assembly were estimated to be higher than the average coolant outlet temperature by 22.8°C, when flow split and heat transfer between sub-channels were considered. Additional sensitivity calculations were performed for the various inter assembly gap flow rates and turbulent flow mixing. The maximum coolant and cladding temperatures, which are major parameters in the conceptual design stage, were not strongly affected by the turbulent mixing under the HYPER design conditions. In addition, some corrosion experiments on static conditions are performed for the core structural materials including beam tube material and fuel cladding.

Figure 9. **Beam target and window design concept**



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 behaviours 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 built in KAERI in order to test corrosive properties of the coolant.

Acknowledgements

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