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The Conceptual Study of Thorium Fuel Cycle for TRU Transmutation and Energy Production

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ABSTRACT

The concept of thorium molten salt fuel cycle has been studied as the alternative fuel cycles for the production of electricity as well as transmutation of TRU elements. At the early stage of the fuel cycle, fissile plutonium isotopes in TRU elements will be incinerated to produce energy and to breed U-233 from thorium. Preliminary calculation showed that periodic removal of fission products and small amount of TRU elements addition could maintain the criticality without separation of Pa-233. At the end of the fuel cycle, the composition of fissile plutonium isotopes in TRU elements was significantly reduced from about 60% to 18%, which is not attractive any more for the diversion of plutonium. The TRU elements remained at the end of fuel cycle can be incinerated in HYPER having fast neutron spectra.

INTRODUCTION

Most of the existing reactors in Korea utilize the low enriched uranium. Since spent fuels resulted from these reactors contain long-lived radionuclides including plutonium isotopes, great attentions are paid how to handle them. Plutonium isotopes should be either completely isolated from the biosphere due to its toxicity, or utilized effectively without any possibility for its diversion to military purpose. Though direct disposal of spent fuels in deep geological repository has been considered, it seems not to remove all the problems. Rather than just covering all of the dangerous possibilities, there must be a safe and more effective way to deal with this notorious waste.

Thus Korea Atomic Energy Research Institute(KAERI) has initiated the

transmutation research for minor actinides and long-lived fission products since 1992. Some feasibility studies were performed and a couple of basic guidelines were introduced to decide the type of transmutation system[1]. An accelerator driven subcritical reactor, named HYPER (Hybrid Power Extraction Reactor) was found to be promising for the transmutation purpose. HYPER is planned to transmute about 380 kg of TRU elements a year and produce 1000 MWth power at 1 GeV and 16mA proton beam. The support ratio of a HYPER to LWR units producing the same power is assumed to be 5 to 6.

The concept of thorium molten salt fuel cycle as a supplementary fuel cycle to HYPER has been studied in this paper. This concept should fulfill the transmutation of long-lived radionuclides, utilization of nuclear material other than U-235 for diversification of energy resources and proliferation resistance. Among various fuel types evaluated for the system, thorium molten salt fuel has been one of the choices for the purpose. The fuel cycle consists of following characteristics: 1) Enrichment facility is not required for the fuel cycle. 2) Plutonium is not separated from other TRU elements. 3) TRU elements separated by pyrochemical processes are mixed with thorium as a fluoride form. And, 4) there is no separation process for protactinium.

RESULTS AND DISCUSSION

Benefit of thorium molten salt fuel.

The advantages of thorium fuel cycle as compared to uranium-based fuel cycle involve a significant reduction in the yield of transuranic actinides, especially plutonium, and production of U-233 during incinerating fissile materials[2]. The U-233 bred by thorium is a superior fissile material for thermal reactors than either U-235 or Pu-239. The thorium fuel cycle has an attractive negative temperature coefficient in thermal reactors that enhance reactor safety. The production of fission products which is the main contributor to reactor poisoning is about 25% less for U-233 than for U-235 or Pu-239. Finally, thorium is an abundant resource than uranium in the earth.

The neutronic properties of each nuclear fuels are described in Table 1[3]. When the number of neutrons produced per neutron absorbed in fissile material is greater than 2.0, it is theoretically possible to generate fissile material at a faster rate than it is consumed. In thermal reactors fueled with plutonium, the number of neutrons produced per neutron absorbed is less than 2.0 and breeding is impossible. For U-233, on the other hand, this number is substantially greater than 2.0, and breeding is practicable in a thermal reactor.

Another advantage of using thorium molten salt fuel is that the separation of plutonium from other TRU elements is not necessary. MOX fuels for PWR and FBR are required to separate plutonium. Plutonium presents a difficult problem in

proliferation resistant because it cannot be simply denatured. It has been known that almost any combination of plutonium isotopes can be made into a weapon unless the Pu-242 content is very large. Thus, pure plutonium separation from spent fuels should be prohibited all the time.

Molten salt fuel cycle was introduced because it gave many advantages[4]. There is no fuel fabrication process and it facilitates to remove the fission products periodically and to provide homogeneous burning of the transmuted materials. The fission products to be removed periodically will be noble gases, seminoble and noble metals.

Preliminary calculations of thorium molten salt fuel.

Preliminary calculation was made by MCNP and ORIGEN code for following conditions. Initial core of the thorium molten salt reactor had 20.9 ton of thorium and 8.89 ton of TRU elements to produce 1,000 MWth power. The average neutron flux at the beginning of cycle was assumed to be $\sim 5 \times 10^{14}$ n.s/cm^2.s. The net multiplication factor at the beginning of the cycle was assumed 1.049. First removal of fission products and addition of 100 kg TRU elements were made after 700 days at the beginning of the cycle. Thereafter every 1,000 days fission products were removed and 80 kg of TRU elements were added. TRU elements addition was necessary to maintain the criticality. The thorium concentration and total inventory of actinides were allowed to decline naturally. The isotopic compositions of TRU elements at the beginning were shown in Table 2 with those at the end of the cycle. As can be seen, fissile plutonium contents in TRU elements were reduced significantly from 59.8% to 17.6%. Total added TRU elements amounted to 9,546 kg and 3,432 kg of TRU elements was remained at the end of the cycle. Thus 6,114 kg of TRU elements were consumed for 32 years which was equal to 190 kg of TRU elements burning every year. For thorium 8,160 kg were consumed and 1,662 kg of U-233 were remained in the used molten salt fuel.

From the calculation, it is obvious that the thorium fuel cycle can have a significant impact on the disposal problem of PWR spent fuels and can effectively utilize fissionable TRU elements to generate new fissile materials from thorium. The TRU elements production level of thorium fuel cycle was lower than that of the uranium cycle. Thus thorium molten salt fuel cycle may be one of the alternative fuel cycles for the transmutation of TRU elements. The TRU elements remained at the end of the thorium molten salt fuel cycle can be incinerated in the system having fast neutron spectra.

Pyrochemical processes for HYPER fuel cycle.

There are many possible processes to separate TRU elements from spent fuels. Because Purex process, though well established, is considered not to be proliferation resistant,

the combination of pyrochemical processes will be employed to separate TRU elements from spent fuels. Since the decontamination factor of pyrochemical processes is not sufficiently high, it is well known that TRU elements obtained by these processes cannot be utilized for military purposes without further purification.

Fig.1 shows the pyrochemical processes for the separation of uranium and TRU elements from PWR spent fuels. After clad materials are removed from spent fuels, either fluorination process or direct oxide reduction process will be applied. If fluorination process is chosen, uranium hexafluoride can be separated easily by its high volatility and be converted to uranium dioxide fuels for CANDU reactors. TRU elements can be separated from fission products by pyrochemical process[5]. Finally it is converted to fluoride forms and mixed with thorium molten salt fuels. If direct oxide reduction process is applied[6], electrorefining process will provide the separation of uranium, fission products, and TRU elements.

Fig.2 briefly shows the concept of flow diagrams of HYPER fuel cycle during operation. Fission products will be removed periodically and thorium, uranium and TRU elements will be re-circulated. No other separation processes such as protactinium separation will be considered.

Proliferation resistance of HYPER fuel cycle.

Proliferation-resistant fuel cycle is defined as one in which at each point of the cycle the fissile material is so degraded that it is not realistic to extract it and use it to produce a fission weapon. In this fuel cycle concept, because the separation of protactinium may be not sufficiently proliferation resistant, the isolation process of protactinium would not be involved [6]. As a result, U-233 is always contaminated by U-232 and its daughter products, some of which are hard γ-emitters. This makes it much more difficult to handle. By contrast, Pu as an α-emitter can be more easily diverted. It has been known that U-233 is an inferior material for arsenal purpose than Pu-239 because nuclear weapon basically depends on fast fissions. U-233 can be easily denatured by the addition of U-238 at the beginning of the cycle if really needed. This can ensure that no weapon's grade uranium is present at any point of the thorium cycle though it is contradicted to the transmutation purpose.

The fissile plutonium isotopic composition at the end of the thorium molten salt fuel cycle is transformed to uninteresting composition as a weapon material. Thus using TRU elements for thorium fuel cycle may be more proliferation resistant than direct disposal of spent fuels.

HYPER fuel cycle for transmutation and energy production.

Depending on the nuclear programs of each country, reactor types and fuel cycles for the transmutation may be different. In Korea where PWR and CANDU are being operated, the study on the utilization of uranium in spent fuels of PWR has been performed and Direct Use of PWR spent fuel In CANDU (DUPIC) program is in progress. In order to utilize the plutonium, reprocessing of spent fuels and fabrication

of MOX fuels for PWR in foreign country are being considered.

Because Korea does not have a reprocessing plant, a combination of pyrochemical processes is being considered to separate TRU elements and long-lived fission products from the PWR spent fuels for ADS purpose. Fig.3 shows a brief concept of HYPER fuel cycle combined with other fuel cycles. The uranium, which is a by-product of the pyrochemical process, would be used for CANDU fuels. And TRU elements will be sent to either thorium molten salt reactor for breeding U-233 or ADS for transmutation. In order to use thorium as a nuclear fuel, a neutron source such as U-235, Pu-239, or an accelerator is necessary to supply enough neutrons. In the thorium molten salt reactor, fissile materials in TRU elements are incinerated to produce neutrons and energy. Some of the neutrons are used to breed U-233 and the other neutrons are used to maintain the criticality of the molten salt reactor. The TRU elements remained at the end of the thorium fuel cycle can be incinerated in HYPER having fast neutron spectra.

SUMMARY

The thorium molten salt fuel cycle could produce energy by incineration of fissile plutonium and U-233 bred from thorium. Periodic removal of fission products and small amount of TRU elements addition could maintain the criticality without separation of Pa-233. TRU elements of 190 kg per year could be burned for 32 years at a plant of 1,000 MWth power with consumption of 8,160 kg thorium. The utilization of thorium diversifies energy resources. Fissile plutonium contents in TRU elements could be reduced from 60% to 18%. No separations of plutonium and Pa-233, and U-233 contaminated by U-232 fulfil the proliferation resistance. The TRU elements remained at the end of fuel cycle can be incinerated in HYPER having fast neutron spectra.

The following studies are needed for the verification of thorium fuel cycle feasibility:

1) Further code calculations for conditions to remove only the noble gases, seminoble and noble metals among fission products and for the determine of the step where the thorium content is kept constant. 2) Effective separation process of fission products from the used thorium molten salt fuel. 3) Oxide removing process from the molten salt fuel. 4) Solubility of TRU elements and fission products in thorium molten salt. 5) Methods to control the oxidation states and chemical activities of TRU elements and fission products in molten salt fuel. And 6) on-line analytical techniques for process control.

ACKOWLEDGMENT

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Table 1. Neutronic properties of each nuclear fuels

Isotope	U-235	Pu-239	U-233
Obtained from	Natural U	U-238	Th-232
Neutron produced per			
- Fission	2.418	2.871	2.492
- Thermal neutron absorbed	1.98	1.86	2.2
Absorption cross section, b			
- Thermal neutrons	555	1618	470
- Fast neutrons	1.5	2	2

Table 2. Isotopic composition of TRU elements at the beginning and the end of thorium fuel cycle.

Nuclide	Weight Fraction (%) at the Beginning of Cycle	Weight Fraction (%) At the End of Cycle
Np-237	4.6	0.9
Pu-238	1.4	9.2
Pu-239	52.1	7.3
Pu-240	23.7	43.0
Pu-241	7.7	10.3
Pu-242	4.5	15.4
Am-241	5.0	4.2
Am-243	0.8	3.9
Cm-244	0.2	4.3

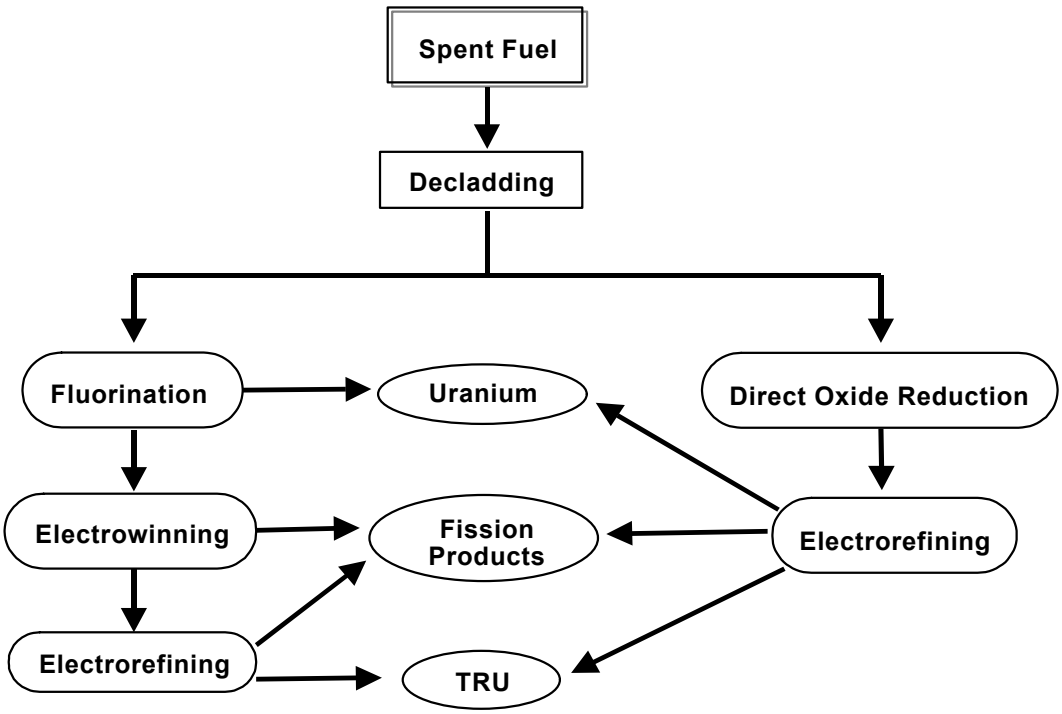

Fig. 1. Pyrochemical processes for HYPER fuel cycle

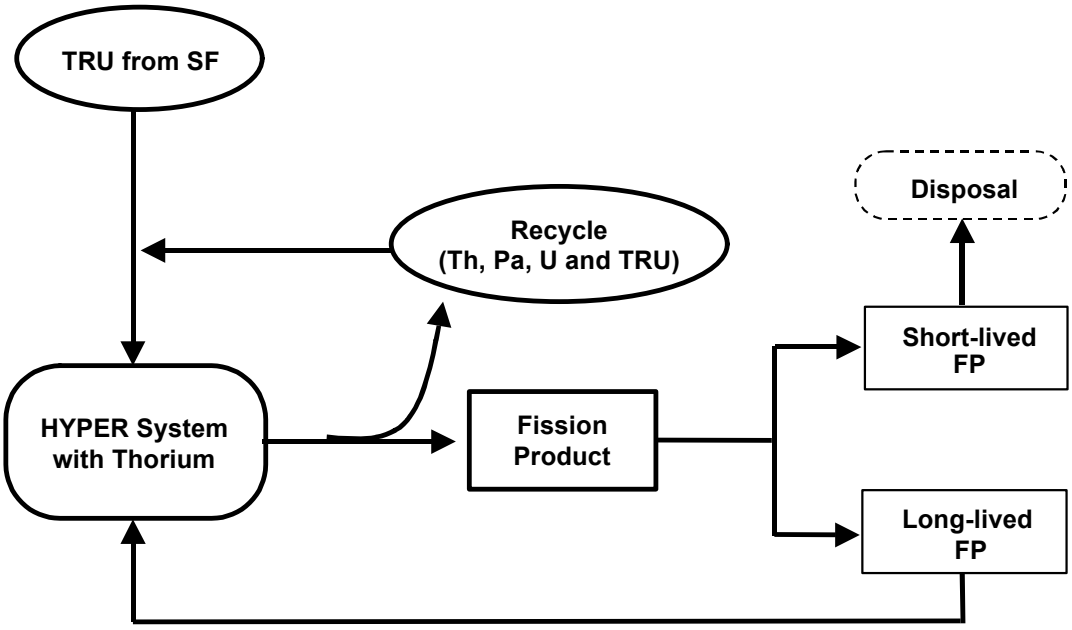

Fig. 2. Thorium fuel cycle in combination with HYPER system

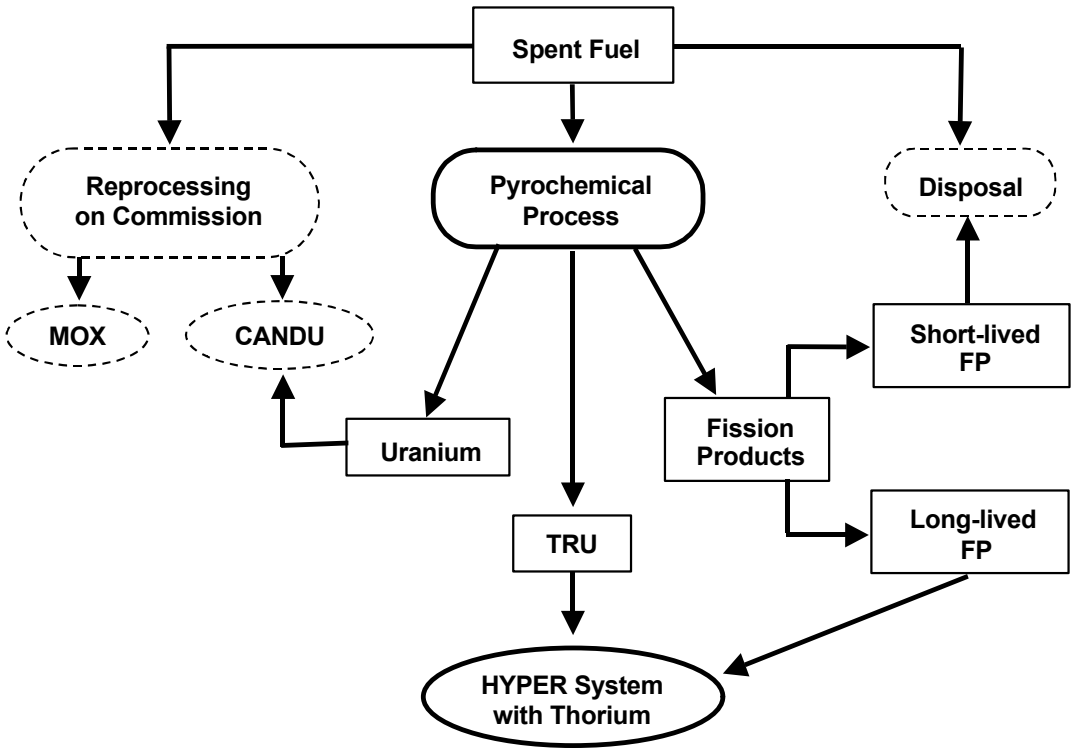

Fig. 3. Pyrochemical process in combination with HYPER fuel cycle