THORIUM FUEL CYCLE CONCEPT FOR KAERI'S ACCELERATOR DRIVEN SYSTEM PROJECT*

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Abstract. Korea Atomic Energy Research Institute (KAERI) has been carrying out accelerator driven system related research and development called HYPER for transmutation and energy production. HYPER program is aiming to develop the elemental technologies for the subcritical system by 2001 and build a small bench scale test facility (~5MW(th)) by the year 2006. Some major features of HYPER have been developed and employed, which are on-power fueling concepts, a hollow cylinder-type metal fuel, and Pb-Bi as a coolant and spallation target material. Another fuel cycle concept for HYPER has been also studied to utilize thorium as a molten salt form to produce electricity as well as to transmute TRU elements. At the early stage of the fuel cycle, fissile plutonium isotopes in TRU will be incinerated to produce energy and to breed ^{233}U from thorium. Preliminary calculation showed that periodic removal of fission products and small amount of TRU addition could maintain the criticality without separation of ^{233}Pa. At the end of the fuel cycle, the composition of fissile plutonium isotopes in TRU was significantly reduced from about 60% to 18%, which is not attractive any more for the diversion of plutonium. Thorium molten salt fuel cycle may be one of the alternative fuel cycles for the transmutation of TRU. The TRU remained at the end of fuel cycle can be incinerated in HYPER having fast neutron spectrums.

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, Korean government has paid great attentions 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 covers all of the dangerous possibilities, there must be a safe and more effective way to deal with this notorious waste.

Thus 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 research is being performed within the frame of the national long term nuclear research plan. The whole development schedule is subdivided into two phases. The design concept of the transmutation system and some basic key technologies are scheduled to be developed in Phase I (1997 – 2001). A small scale test facility (~5MW(th)) is to be designed and built in Phase II (2002 –2006). 1 GeV 16mA proton beam is designed to be provided for HYPER. HYPER is planned to transmute about 380 kg of TRU a year and produce 1000 MW(th) power. The support ratio of HYPER to LWR units producing the same power is assumed to be 1 to 5 ~6.

First of all, the fuel cycle concept was focused on transmutation of long-lived radionuclides, thorium utilization 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:

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* 1998 meeting.

- Enrichment facility is not required for the fuel cycle.
- Plutonium is not separated from other TRU elements.
- TRU elements separated by pyrochemical processes are mixed with thorium as a fluoride form.
- There is no separation process for protactinium.

WHY THORIUM MOLTEN SALT FUEL IS BENEFICIAL?

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 ^{233}U during incinerating fissile materials [2]. The ^{233}U bred by thorium is a superior fissile material for thermal reactors than either ^{235}U or ^{239}Pu. 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 ^{233}U than for ^{235}U or ^{239}Pu. Finally, thorium is an abundant resource than uranium. The neutronic properties of each nuclear fuels are described in Table I [3].

Table I. Neutronic properties of each nuclear fuels

Isotope	^{235}U	^{239}Pu	^{233}U
Obtained from	Natural U	^{238}U	^{232}Th
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

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 ^{233}U, 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 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 ^{242}Pu 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 to produce 1,000 MW(th) 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 100 kg of TRU addition 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 were added. TRU 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 at the beginning were shown in Table II with those at the end of the cycle.

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

Nuclide	Weight Fraction (%) Beginning of Cycle	Weight Fraction (%) 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

As can be seen, fissile plutonium contents in TRU were reduced significantly from 59.8% to 17.6%. Total added TRU amounted to 9,546 kg and 3,432 kg of TRU was remained at the end of the cycle. Thus 6,114 kg of TRU were consumed for 32 years which was equal to 190 kg of TRU burning every year. For thorium 8,160 kg were consumed and 1,662 kg of ^{233}U 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 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. The TRU remained at the end of the thorium molten salt fuel cycle can be incinerated in the system having fast neutron spectrums.

PYROCHEMICAL PROCESSES FOR HYPER FUEL CYCLE

There are many possible processes to separate TRU 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 from spent fuels. Since the decontamination factor of pyrochemical processes is not sufficiently high, it is well known that TRU 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 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 can be separated from fission products by pyrochemical processes [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 will be re-circulated. No other separation processes such as protactinium separation will be considered.

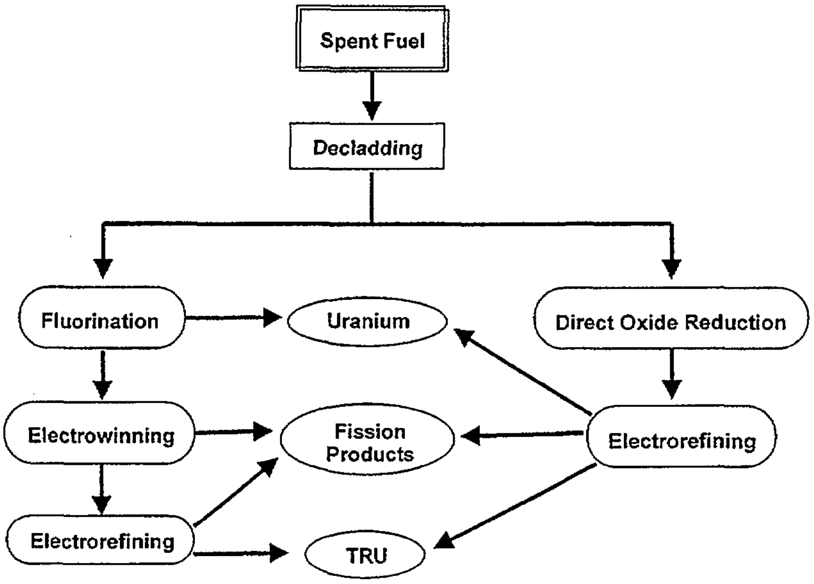

Figure 1. Pyrochemical processes for HYPER fuel cycle.

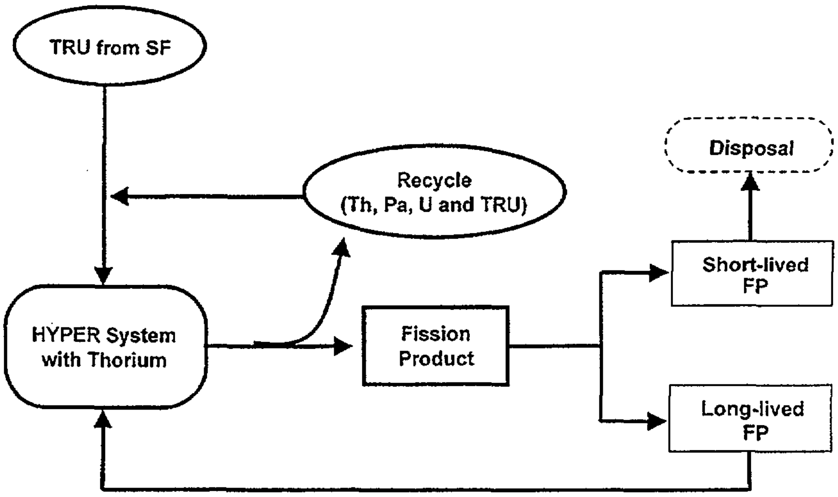

Figure 2. Fuel cycle for HYPER system.

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, ^{233}U is always contaminated by ^{232}U 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 ^{233}U is an inferior material for arsenal purpose than ^{239}Pu because nuclear weapon basically depends on fast fissions. ^{233}U can be easily denatured by the addition of ^{238}U 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 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 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.

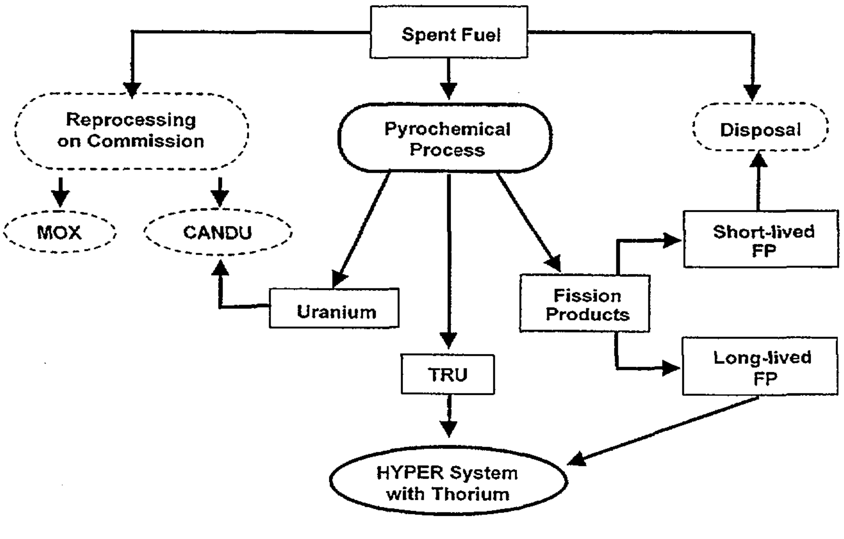

Figure 3. Concept of HYPER fuel cycle.

The uranium, which is a by-product of the pyrochemical process, would be used for CANDU fuels. And TRU will be sent to either thorium molten salt reactor for breeding ^{233}U or ADS for transmutation. In order to use thorium as a nuclear fuel, a neutron source such as ^{235}U, ^{239}Pu, or an accelerator is necessary to supply enough neutrons. In the thorium molten salt reactor, fissile materials in TRU are incinerated to produce neutrons and energy. Some of the neutrons are used to breed ^{233}U and the other neutrons are used to maintain the criticality of the molten salt reactor. The TRU remained at the end of the thorium fuel cycle can be incinerated in HYPER having fast neutron spectrums.

FUTURE DEVELOPMENT PLAN

- Further code calculations will be made for following conditions:
  * Only the noble gases, seminoble and noble metals among fission products are removed.
  * Where the thorium content is kept constant.
- Effective separation process of fission products from the used thorium molten salt fuel will be evaluated.
- Oxide removing process from the molten salt fuel will be studied.
- Solubility of TRU and fission products in thorium molten salt will be measured.
- Methods to control the Redox and activity conditions of molten salt fuel will be studied.
- On-line analytical techniques will be developed.

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