

Conceptual Study of Actinide Reformer Using High-power Proton Accelerator

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In Japan, the nuclear fuel cycles based on FRs will be established. To realize the fuel cycle, however, americium (Am) and curium (Cm) should be carefully managed because they may affect the soundness of nuclear fuels if they are recycled with plutonium. On the other hand, if we dispose of Am, they will affect the design of the geological repository because of their long half-lives. We propose a new system concept called an "Actinide Reformer", whose purpose is the effective transmutation of Am and Cm. In the concept, Am and Cm are separated from the spent fuel by the advanced aqueous reprocessing and converted to chloride molten-salt fuel on which we have many experiences in the development of the dry reprocessing. Using such liquid-type fuel, many defects on Am and Cm such as high radioactivity and heat release and chemical incompatibility can be overcome. The Actinide Reformer is designed as an accelerator-driven system which consists of a 10MW-class cyclotron, a tungsten target and a subcritical core. Molten-salt fuel is reprocessed to extract fission products and to add Am and Cm. The system is designed to recover overall electricity to drive system itself during the lifetime of the reactor.

I. INTRODUCTION

Management of radioactive wastes from nuclear power plants is one of the important issues for sustainable nuclear power generation. To propose the way to reduce the environmental impact from disposal of high-level radioactive wastes and to improve the cost and space efficiency of geological disposal site, the long-term research and development program for Partitioning-Transmutation (P-T) technology using accelerator-driven system^[1] has been performed. In the concept of transmutation by a dedicated accelerator-driven system (ADS), minor actinides (MA) are separated by the four-group partitioning process and fabricated to the pellet nitride fuel. However, handling americium (Am) and curium (Cm) and their daughter nuclides poses difficulties even in the P-T fuel cycle because of their high decay heat and radioactivity. These nuclides also give the

chemical instability, namely the low vapor pressure of Am, which harms the soundness of burnup fuel pellet properties. In contrast, neptunium (Np) has similar chemical properties as uranium (U) and plutonium (Pu) and shows good compatibility not only in nitride fuel but also in oxide fuel. There are no significant difficulties to handle Np in a MOX based fuel cycle.

To perform transmutation of MA effectively, a fast fission reaction is desirable. However, because neutron capture reaction occurs simultaneously in a fast spectrum core, large MA loadings and high thermal output are required for efficient transmutation. Consequently, the currently proposed accelerator-driven transmutation systems must be connected to a high intensity proton linear accelerator.

Considering above mentioned characteristics of MA and dedicated transmutation systems, we propose a "Actinide Reformer" concept which is a revised molten-

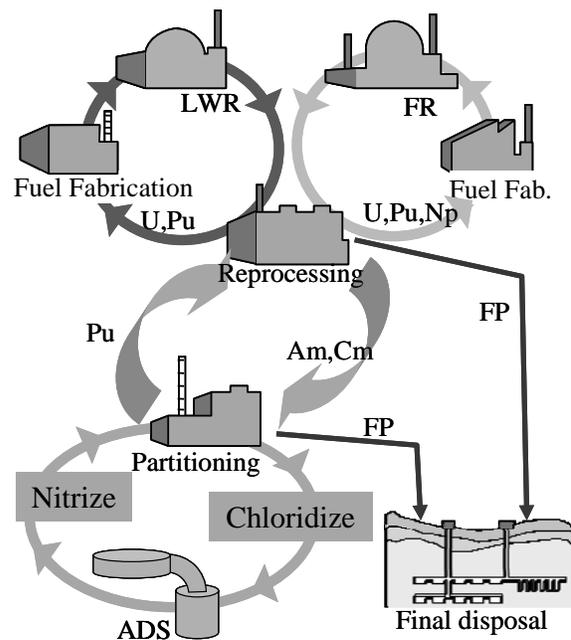


Fig. 1 Actinide Reformer Fuel Cycle Concept

salt ADS. There are two concepts for actinide reformer, one reforms Am and Cm into Pu by neutron capture reactions and decay to reduce the burden in the fuel cycle. The other is more dedicated system to burn MA in a small closed fuel cycle. In this paper, outline of the both concepts of actinide reformer, neutronic performance including burnup analysis and preliminary mass balance by introduction of the actinide reformer are described.

II. OUTLINE OF ACTINIDE REFORMER

Current LWR reprocessing process (PUREX process) aims at recovering U and Pu from spent nuclear fuel. Through the process, Am and Cm are easily separated from other actinides. However, to separate Np, whose chemical properties are close to Pu, requires an additional recovering step within the PUREX process. Because of the similarity of chemical and neutronic performance of Np and Pu, it is not so difficult to treat them identically in the MOX fuel based nuclear fuel cycle and it is also effective for reduction of cost and secondary waste.

The fuel cycle based on actinide reformer concept is illustrated in Fig. 1. When the actinide reformer fuel cycle is adopted, Np in the spent fuel from power reactors is recovered and recycled with Pu. Omission of Np separation process from current PUREX reprocessing process may give more cost effectiveness, safety and secondary waste reduction. From the reprocessing process, Am and Cm are sent to the actinide reformer cycle. At first, Am and Cm are transformed to chloride molten-salt fuel. By using Am and Cm as a molten-salt fuel form, facilities such as fuel fabrication, storage, assembling/disassembling, etc. are not necessary. It is also needless to consider the engineering limitations of decay heat removal, linear power and chemical instability in solid fuel form. There also arise some demerits of liquid fuel: high radioactive nuclides spreads whole primary cooling circuit, risk of FP sedimentation formation and irradiation effect to chemical binding and so on.

For effective burning of molten-salt fuel, actinide reformer is designed as an ADS which is composed of a 10MW-class cyclotron, a tungsten spallation target and a molten-salt fuelled subcritical core. The engineering feasibility of the accelerator and the spallation target can be set in the range of the existing technology. In the viewpoint of neutronics, application of liquid fuel for subcritical system gives many advantages such as improved neutron economy, passive safety by fuel density effect, applicability of on-line adjustment of fuel composition. These neutronic characteristics are also useful to construct subcritical core. To compensate the demerit of liquid fuel, simple and small primary loop is

considered. The power from fission reactions of actinides is recovered and used for electricity to operate its own accelerator. For reprocessing of spent molten-salt fuel, existing PUREX process and four group partitioning process, which is proved by laboratory scale, can be applied. By using four group partitioning process together with PUREX process, spent fuel is roughly classified to minor actinides (mostly remaining Am and Cm), reformed Pu and several fission product groups. The group containing MA is recycled in the actinide reformer cycle. In Pu recovering case, recovered Pu is returned to the power reactor fuel cycle and used for power generation. Effect of inclusion of reformed Pu into power reactor MOX fuel will be discussed in the following section.

III. CHARACTERISTICS OF PLUTONIUM RECOVERING SYSTEM

III.A Initial Core Configuration

To determine an optimum configuration of subcritical core dimensions and initial fuel composition, neutronic analysis of the subcritical core was performed. In the parametric survey to specify the core dimensions, the initial k-effective value (k-eff) is fixed around 0.98. It is also considered that the initial MA inventory is limited to less than 2 tons to transmute MA effectively. Because of the rather high melting point of molten-salt, the spallation target is separated from the molten-salt fuel tank. This configuration is useful to reduce the coolant temperature of the target in order to improve the structural integrity of the beam window and use water as coolant material. This spallation target configuration also allows exchanging the target without any fuel handling procedures. The radius of the spallation target is specified from a limitation of the proton beam current density, which is tentatively set to $20\mu\text{A}/\text{cm}^2$. Using this beam current density limitation and the corresponding spallation target radius, maximum allowable proton beam power is set to 5 MW with proton energy of 800 MeV. Chloride molten-salt is loaded into the fuel region. The actinide-based molten-salt $[(\text{AmCm})\text{Cl}_3]$ is mixed with 10% of lead-chloride $[\text{PbCl}_2]$ to reduce the fission power density. Composition of actinides used in this calculation is 87.7 % of Am and 12.3 % of Cm. A stainless steel reflector is located around the fuel region to improve neutron economy and radiation shielding performance. The overall size of the subcritical core is about 120cm diameter and 140 cm length. A two-dimensional cylindrical calculation model for the final core configuration is illustrated in Fig. 2.

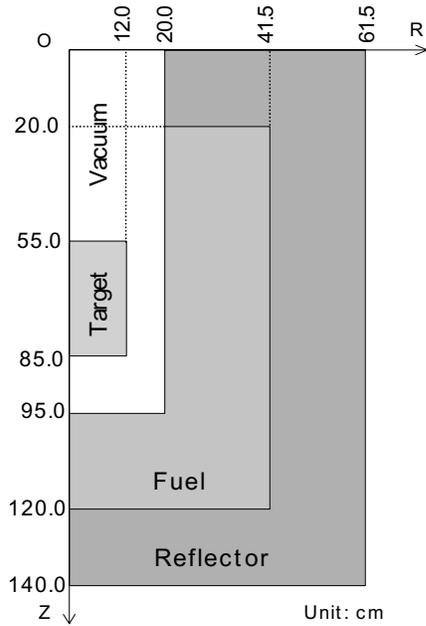


Fig.2 Neutronic Analysis Model

Table 1 summarizes the analysis results obtained by using MCNPX-2.5 code^[2] and JENDL-3.3 cross section library^[3]. An initial k_{eff} of the subcritical core is determined about 0.98 with initial actinide inventory of 1,625 kg. Average power densities of the target region and the subcritical core region are 370 W/cm^3 and 380 W/cm^3 , respectively. The power peaking factor, which usually gives rather higher value for subcritical system than for a critical reactor, has no limitation for liquid fuel. By the injection of rated power (800 MeV - 5 MW) proton beam, thermal output from subcritical core reaches around 170 MW. Fig. 3 shows the average neutron energy spectrum in the subcritical core region. Because of the

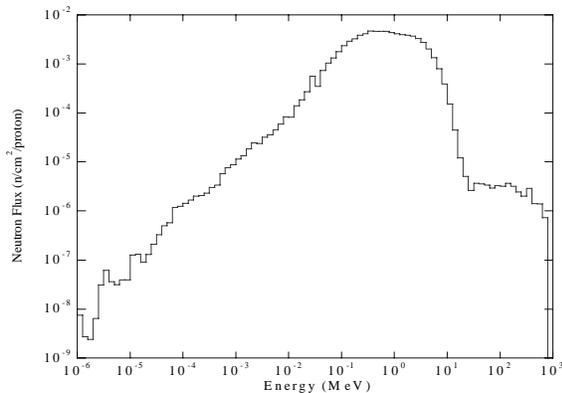


Fig.3 Average Neutron Energy Spectrum in Core

application of chloride molten-salt to the fuel, a hard neutron energy spectrum similar to that of fast reactor is obtained.

Table 1 Neutronic Analysis Results for Initial Core

Target Radius / Height [cm]	12.0 / 30.0
Target Material / Coolant	Tungsten / Water
Fuel Zone Radius / Height [cm]	41.5 / 100
Fuel Density [g/cm^3]	5.70
MA Fraction (Am:Cm)	88 : 12
Fuel Fraction ($\text{MACl}_3\text{:PbCl}_2$)	90 : 10
Proton Energy [MeV]	800
Proton Beam Power [MW]	5
Initial $k_{\text{effective}}$	0.9802
Thermal Output [MW]	170
Av. Power Density (Target) [W/cm^3]	368
Av. Power Density (Core) [W/cm^3]	380
Av. Proton Beam Current [$\mu\text{A/cm}^2$]	19.9
Initial MA Loading [kg]	1625

III.B Burnup Analysis

Using the initial core configuration mentioned in the previous section, burnup analysis was performed. To apply the simulation of on-line refueling and fuel concentration adjustment for the burnup calculation, ABC-SC code system^[4] and JENDL-3.3 based cross section library were used. Decontamination Factor (DF) of actinides (U, Np, Pu) and FP (Fission Products) excluding rare earth elements were specified to 100 (99 % recovery). DF for rare earth is assumed to be 2 (50 % recovery). For this preliminary analysis, burnup period is set to 300 Equivalent Full Power Day (EFPD) with 65 day cooling period.

Fig. 4 summarizes the removed nuclides amount for each burnup cycle. From the contents of extracted nuclides, the equilibrium burnup cycle is established after the seventh cycle. By the definition of these analysis conditions, about 100 kg of Am and Cm can be fed to one actinide reformer annually at equilibrium cycle and then, 50.8 kg of FP and 42.3 kg of reformed actinides (Np and Pu) are recovered simultaneously. To support the Rokkasho-scale reprocessing plant (430 kg of Am and Cm are annually recovered), for example, 5 units of actinide reformers are required. Using 5 units of actinide reformers, 500 kg of Am and Cm can be loaded in actinide reformer fuel cycle. And then, total nuclide amount which must be disposed of into the final disposal site can be decreased. About 212 kg of Np and Pu are also recovered and it can be used as a fuel for power generation.

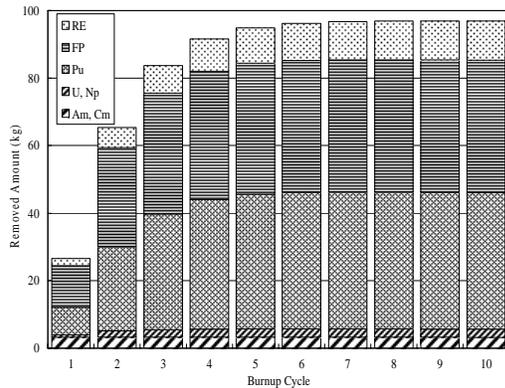


Fig.4 Removed Nuclides within each Burnup

III.C Impact of Reformed Plutonium on Power Reactor Fuel Cycle

In the result of previous burnup analysis, the isotope composition ratio of reformed Pu is 57 % of Pu-238, 30 % of Pu-240 and 13 % of Pu-242. In this section, the influence of reformed Pu usage in power reactor fuel cycle is discussed.

Table 2 summarizes the typical Pu concentration of MOX fuels considered for both LWR and FR^[5]. The table indicates that the FR-MOX with MA recycle case, which loads about 5 % of MA in MOX fuel, gives higher concentrations of Pu-238 and Am-241. In the cases of the actinide reformer cycle, Am and Cm are sent to actinide reformer and reformed to Pu that is to be loaded into the FR. The Pu loading amount in LWR-MOX case are not so large even adding the reformed Pu into LWR-MOX fuel. In the case of FR-MOX without MA recycle, it is also similar to LWR-MOX case. In contrast, the FR-MOX with MA recycle case, which transmutes all MA in its fuel cycle and does not require the actinide reformer concept, the increasing ratio of Pu-238 and overall Pu yields much higher decay heat values than the other cases.

Fig. 5 shows the comparison of fuel decay heat. Applying actinide reformer for LWR and FR without MA recycle case, decay heat is not significantly increased. In contrast, FR-MOX with MA recycle case, MA is recycled together with Pu in fast power reactor, shows large influences caused by Cm-244. A similar characteristic arises for background neutron emission. In the viewpoint of power generation cost, it is desirable to handle more simple fuel like FR-MOX without MA recycle case. By applying actinide reformer cycle, Cm-244 is confined in the actinide reformer cycle and reformed Pu gives rather small impact to the power reactor fuels.

Table 2 Isotopic Composition of Pu in Various Reactor

Nuclides (tons/year)	Concepts		
	LWR-MOX fuel	FR-MOX fuel without MA recycle	FR-MOX fuel with MA recycle
Pu loading (incl. MA)	4.6 %	19.7 %	21.7 %
²³⁸ Pu	0.19	0.24	0.70
²³⁹ Pu	3.85	26.07	26.40
²⁴⁰ Pu	1.12	12.50	13.21
²⁴¹ Pu	1.15	1.19	1.21
²⁴² Pu	0.66	1.23	1.65
²⁴¹ Am	0.06	0.29	1.52
Others	0.0	0.0	1.08

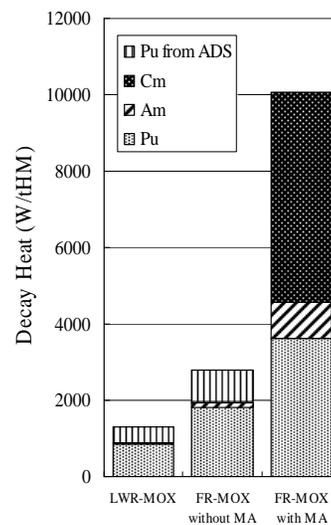


Fig.5 Comparison of Released Decay Heat

IV. DEDICATED TRANSMUTATION SYSTEM

IV.A Outline and Neutronic Characteristics

The first concept of actinide reformer aims at reforming the harmful actinides, Am and Cm to useful actinides, Pu. However, there still exists rather small impact on commercial nuclear fuels and several complicated process is required to reprocess the spent molten-salt fuel. If the actinide reformer cycle is completely separated from power generation fuel cycle, there is no significant impact to power generation cycle. In this chapter, dedicated molten-salt fuel transmutation system focused on transmutation of Am and Cm is considered.

In this case, because the remaining actinides in spent molten-salt fuel can be commonly extracted, the dry

reprocessing method can be applicable. The possibility of combination with dry-reprocessing will be discussed in section III.B.

One of the merits for the liquid fuel is a flexible reactivity control by adjusting the fuel level. This capability is more efficient in ADS than in critical systems. By adjusting the subcriticality level from cycle to cycle by the fuel level adjustment, engineering requirement and construction cost of the accelerator can be reduced. In ordinary case of actinide burner, fissile material for fast system, Pu, is produced at early burnup cycles and at the equilibrium condition, k-eff reaches higher value than that of early burn up stages without any compensation. In the case of solid fuel, there are limited methods to control reactivity and then, in JAEA, Pu is added into the start-up fuel to compensate for burnup reactivity loss. For the effective transmutation of actinides, burn up analysis with fuel level adjustment was applied.

Fig. 6 shows the two-dimensional analysis model. The common specification of previously mentioned spallation target was applied. In this case, the height of cylindrical core is selected as a parameter for adjusting k-eff.

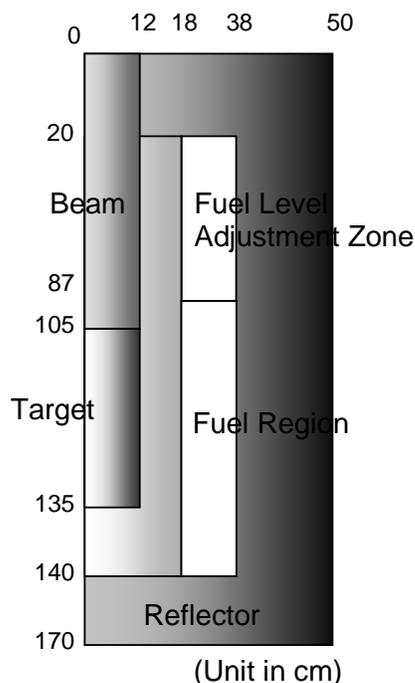


Fig.6 Two-dimensional Analysis Model

Burnup analysis was performed with analysis conditions of 170 MW constant power, 300 day burnup per cycle and 300 day cooling including fuel reprocessing. In the fuel reprocessing process, all fission products are separated and same amount of Am and Cm is

added. In this analysis, fuel level only at the beginning of cycle (BOC) was adjusted.

It is also considered that such liquid fuel must be protected from critical accident by geometrical limitation. In this study, the delicate fuel level adjustment using large fuel tank is not considered and adjustment at early burn up cycles were done by changing the fuel tank height from cycle to cycle to suppress the critical accident. The chloride molten-salt may have limited compatibility with typical structural materials such as SUS316, SUS310S, Inconel 625 and Incoloy 825, fuel tank might be replaced at each burn up cycle^[6,7] to protect a fuel leakage accident by corrosion of structural materials.

Fig. 7 indicates a burnup trend of k-eff with fuel level adjustment. The reactivity change during the burnup is not flattened because on-line purification is not considered. The highest k-eff is tentatively set around 0.97 but it can be changed by tradeoff with actinide loading amount related to reprocessing capacity. The reactivity swing at equilibrium cycle is slightly larger because of the decrease of fuel volume. For the future improvement, on-line reactivity adjustment must be included. It is also important to improve the safety against the critical accident, which increasing the reactivity at the first burn up cycle must be compressed by on-line reactivity adjustment simultaneously.

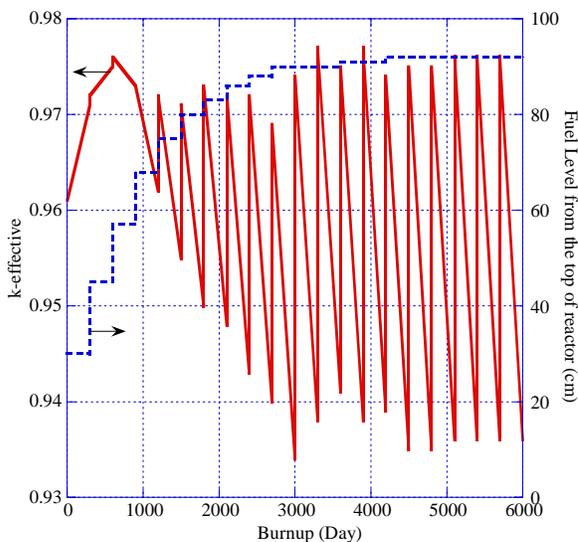


Fig. 7 Trends of Burnup Reactivity and Fuel Level

IV.B Reprocessing of Spent Molten-salt Fuel

In this study, the system adopts the batch refueling manner. From the neutronic analysis result, initial fuel loading is about 1,460 kg (1,300 kg of Am and 180 kg of

Cm) in the core region. The burnup fuel is 1,389 kg of actinides (Pu: 11kg, Am: 1,181kg, Cm: 197kg, and others) and 58 kg of FP. At the equilibrium cycle, about 670 kg of actinides (Am: 405 kg, Cm: 106 kg, Pu: 147 kg) are loaded in the core region. These actinides are transformed into 551 kg of actinides (Am: 355kg, Cm: 107 kg, Pu: 81 kg, and others) and 56 kg of FP. To load about 430 kg of Am and Cm, 8 units are required and about 13 ton of actinides, with consideration of the inventory in the piping and heat exchanger, must be reprocessed at the equilibrium cycle annually.

The design study for dry reprocessing plant that can accept the high level waste from Rokkasho reprocessing plant class facility was performed at CRIEPI^[8]. In the report, the plant was designed to accept about 25 mole of MA per batch, 4 batches per day, and 200 days operation per year. About 200 liter of chloride salt (LiCl-KCl) was used per one batch. In such case, the annual amount of processed MA is about 5 tons and then, at least two processing lines are required for the actinide reformer cycle. However, no processes are required for fuel disassembling, cutting, fabrication and re-assembling. Since MA is already chlorinated, the denitration and chlorination before the application of extraction processes can also removed. From the cost estimation of the plant, the cost of equipments decreases about 30% by eliminating these devices. After the reprocessing, a cadmium recovering process and MA re-chlorination process are required and there are no needs to add fuel re-fabrication processes.

V. CONCLUSION

To improve the safety, cost efficiency and waste reduction, two actinide reformer concepts are proposed. The actinide reformer aims at extracting Am and Cm from a current PUREX process based fuel cycle and reforms them into Pu which has more chemical stability and safety in MOX fuel. The parameter of the components for actinide reformer is set to the feasible values from the viewpoint of current engineering experiences. By adding the actinide reformer concept to the current power generation cycle, fuels for power reactor can keep conventional fuel composition. It is useful to improve safety and cost efficiency of electric power generation.

From the preliminary analysis results, there is a possibility to make actinide reformer system by self-standing systems to reduce the environmental impacts by MA. It is also found that dedicated actinide burner system reflecting actinide reformer concept also gives fairly good transmutation performance. In the latter case, adoption of lower k-eff gives less reprocessing amount than that of higher k-eff by tradeoff of requirement of proton beam power. Detail optimization must be performed by

reflecting the reliability of existing high power accelerators.

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