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EFFECTS OF REPROCESSED URANIUM MULTI-RECYCLE ON THE PROLIFERATION RESISTANCE OF PLUTONIUM AND URANIUM

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ABSTRACT
Effects of reprocessed uranium (RepU) multi-recycle on the proliferation resistance (PR) of Pu and U has been studied. The non-fissile isotope ^{236}U is generated from ^{235}U during burnup in the reactor. The typical isotope ratio of ^{236}U to the remaining ^{235}U isotope in the RepU from used nuclear fuel in light water reactors varies from 0.4 to 1.0, depending on the initial enrichment and the burnup. During burnup, by further absorbing neutrons, some of the ^{236}U atoms become ^{238}Pu via ^{237}Np/ ^{238}Np, which has strong PR because of the high decay heat and spontaneous fission neutron emission. During the centrifuge re-enrichment of ^{235}U in the RepU, ^{236}U is also re-enriched together with ^{235}U. The ratio of ^{236}U to ^{235}U after re-enrichment of the RepU is roughly from 0.3 to 0.4 up to 5% ^{235}U enrichment. Hence, the centrifuge re-enriching of the RepU makes it difficult to produce weapons-grade highly-enriched ^{235}U because ^{236}U is also enriched at the same time. After the re-enriched RepU is burned again in the reactor, naturally ^{235}U is reduced by fission to release energy during the burning and the content of ^{236}U is further increased compared to that after the first cycle. The content of ^{238}Pu is also further increased compared to that after the first cycle, which means the PR of Pu is further increased after the burnup of re-enriched RepU. By repeating this process, the production of highly enriched weapons-grade U becomes more difficult from the multi-recycle RepU. ^{236}U in RepU is an attractive isotope to protect Pu and U from adversarial use. Recycle of minor actinides for protected Pu production (P^3), proposed earlier by Saito elsewhere, and the recycle of the RepU proposed here are both useful and important for the peaceful uses of nuclear energy.

INTRODUCTION
As of 2018, about 290,000 tons of used nuclear fuel has been discharged from nuclear power plants worldwide, of which about 100,000 tons has been reprocessed [1]. Uranium in used nuclear fuel accounts for about 95 wt% of the total weight, so if all the uranium is recovered by reprocessing, about 275,500 tons of uranium will be reusable. The recovery of uranium from used nuclear fuel will contribute to the conservation of uranium resources by reusing the recovered uranium (also known as reprocessed uranium (RepU)) as reactor fuel and will also reduce radioactive waste. The OECD/NEA (Organization of Economic Co-operation and Development/Nuclear Energy Agency) surveys of reusable secondary fission materials excluding natural uranium are summarized in Table 1[2]. At the end of 2005, the total amount of RepU accumulated was about 45,000 tons over the past decade (before 2005), which is equivalent to about 50,000 tons of natural uranium. This is equivalent to supplying the fuel to 1,000 MWe light water reactors (LWRs) of current generation operating at 80% load factor for 300 years, which is almost comparable to Pu in energy volume.

In addition to RepU, secondary fissile materials such as Pu and minor actinides (MA) have also accumulated worldwide in the wake of nuclear electricity production. Since accumulation of these materials has potential risks to threaten nonproliferation and environmental protection. Their build up is essential for making policy and establishing strategy for future nuclear fuel cycles.

To allay the fear of the diversion of Pu to nuclear weapons in the future, the basic concept of "Protected Plutonium Production (P^3) by the transmutation of minor actinides (MA)" has been proposed by Saito et al. [3,4] to increase the proliferation resistance (PR) of Pu accumulated in the used nuclear fuel discharged from LWRs and fast breeder reactors (FBRs). As shown in figure 1, ^{238}Pu present in Pu decays through spontaneous fission producing neutrons at the rate of

$2.6x10^3$n/g/s to deteriorate the nuclear explosive quality of Pu. In addition, the high decay power of ^{238}Pu (570 W/kg) makes the processes of nuclear weapon manufacturing and maintenance technologically difficult. Therefore, the PR of Pu can be improved by enhancing the ^{238}Pu concentration. To denature Pu by enhancing its ^{238}Pu concentration, transmutation of MA can be sought through two nuclear chains from ^{237}Np through ^{238}Np to ^{238}Pu and from ^{241}Am through ^{242}Am and ^{242}Cm to ^{238}Pu as shown in figure 2.

Table 1. Inventory of separated reusable secondary fission materials at the end of 2005 [2].

Material	Quantity (t heavy metal)	Natural uranium equivalent (t U)	Reactor Years of Supply*
Ex-defense HEU	230	70,000	420
Ex-defense Pu	70	15,000	90
Pu	370	60,000	380
RepU	45,000	50,000	300
Enrichment tails	1,600,000	450,000	2,650

* Based on a 1000-MWe LWR operating at an 80% load factor

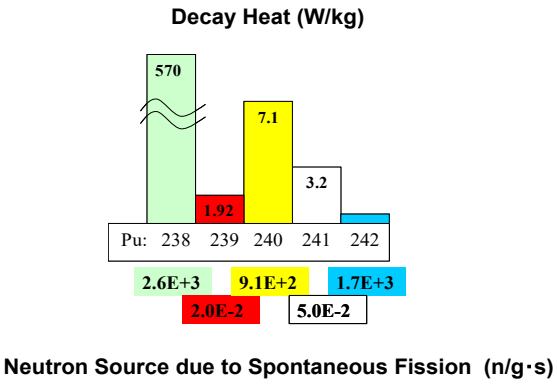

Figure 1. Proliferation resistance attribute of Pu.

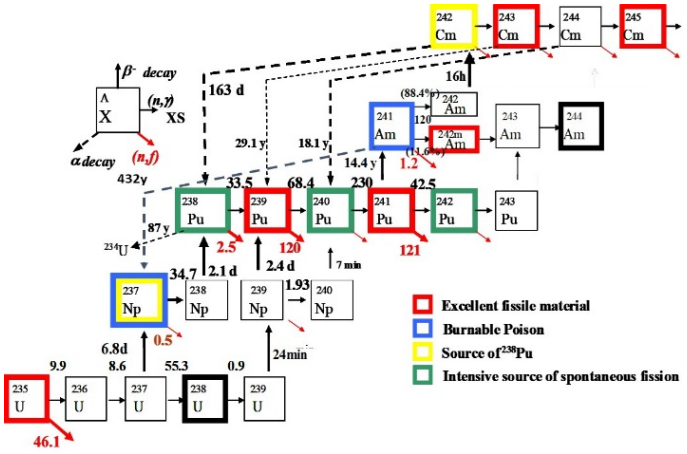

Figure 2. Mechanism of protected Pu production.

The P^3 mechanism by the transmutation of MA has been studied for both thermal and fast reactors [5,6,7,8,9,10,11,12,13,14]. The studies on the P^3 mechanism have been extended to the denaturing of the reactor-grade Pu by the doping mixed oxide (MOX), uranium-free inert matrix (IMF), and thorium dioxide (ThO_2) nuclear fuels in light water reactors (LWRs) with MA [15,16]. In order to validate the P^3 mechanism, two irradiation tests were performed in the experimental fast reactor (JOYO) in Japan and in the advanced test reactor (ATR) in Idaho National Laboratory [17]. To evaluate the PR of Pu, a function, "attractiveness (ATTR) of Pu for nuclear weapon" was proposed as a ratio of potential of fission yield to characteristic of technical difficulty for converting Pu to nuclear explosive device [13, 18]. Attractiveness of Pu decreases dramatically by even-mass-number Pu isotopes doping, because the technical difficulty is enhanced by this type of doping due to the increase in spontaneous fission neutron rate, decay power, and radiation dose rate.

In addition to the protection of Pu by the transmutation of MA, the ^{238}Pu concentration can be enhanced from ^{236}U in the RepU through ^{237}Np/^{238}Np, as shown in figure 2. During the centrifuge re-enrichment of ^{235}U in the RepU, ^{236}U is also re-enriched together with ^{235}U. Since ^{236}U is non-fissile uranium in a thermal reactor, ^{236}U in the RepU has the potential to increase the PR of U in addition to Pu.

In this paper, another innovative way is proposed to increase the PR of U and Pu by the multi-recycle of RepU in LWRs for the promotion of the peaceful uses of nuclear energy in the future.

CRITICAL MASS OF URANIUM

The bare critical mass (in kg) of a metallic U sphere for various isotopes is shown in Figure 3 [19]. The critical mass of ^{235}U used for nuclear weapons is about 45 kg. The bare critical masses of ^{232}U and ^{233}U that do not exist naturally are about 1/3 of that of ^{235}U. Figure 4 shows the bare critical masses of mixed U isotopes where ^{235}U is mixed with ^{238}U or ^{236}U [19]. From figure 4 one can note that U with a mixing rate (enrichment) of ^{235}U to ^{238}U of 20% or less is good for civilian use because it is too heavy for nuclear weapons.

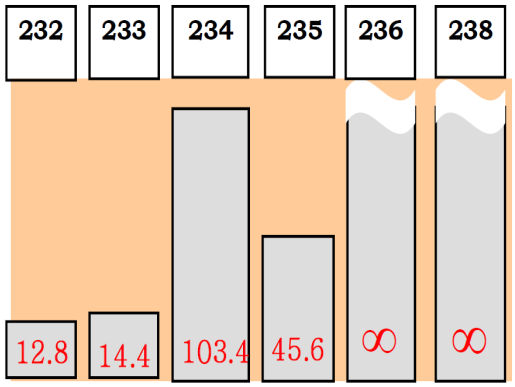

Figure 3. Bare critical mass (kg) of metallic U Isotopes [19].

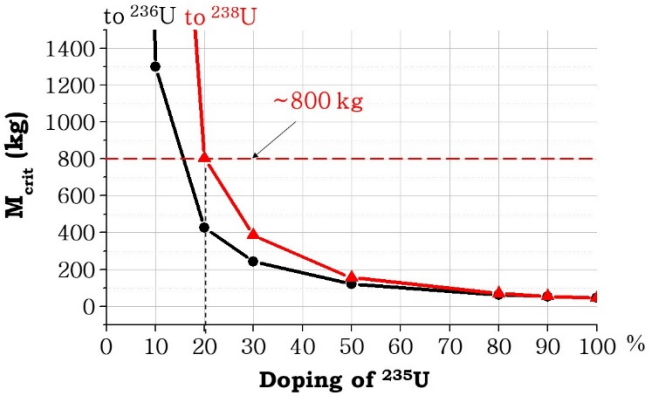

Figure 4. Changes bare critical mass by the mixing of ^{235}U with ^{238}U or ^{236}U [19].

On the other hand, the change in the bare critical mass of U when mixed with ^{232}U, ^{233}U, and ^{234}U to ^{238}U, is shown in figure 5 compared to the case of ^{235}U [19]. The mixing ratio equivalent to the bare critical mass in the case of a mixing ratio (enrichment) of 20% ^{235}U is about half (11.5%) for ^{232}U and ^{233}U. As shown in figure 3, the bare critical mass of ^{234}U is about 2.3 times larger than that of ^{235}U. The mixing ratio equivalent to the bare critical mass at 20% (enrichment) ^{235}U is about 54% for ^{234}U.

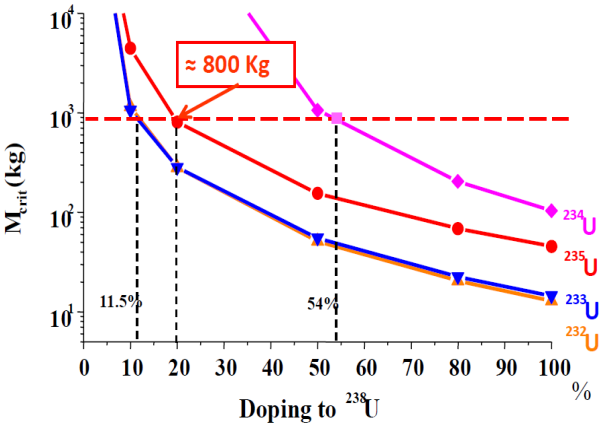

Figure 5. Changes in bare critical mass (kg) by mixing ^{232}U, ^{233}U, ^{234}U, ^{235}U to ^{238}U [19].

REPROCESSED URANIUM FROM LWR and AGR

The non-fissionable isotope ^{236}U is generated from ^{235}U during the fuel burnup in the reactor. The typical isotope ratio of ^{236}U to the remaining ^{235}U isotope in the used fuel in LWR is roughly 0.4 to 1.0 depending on the burnup level for a given initial ^{235}U enrichment in the fresh fuel [20]. Typical isotopic ratios of ^{236}U to the remaining ^{235}U isotope in the used fuel of advanced gas-cooled reactor (AGR) and LWR increase with the burnup as shown in figure 6 [20,21]. The isotopic ratio of ^{236}U to the remaining ^{235}U after the irradiation of the re-enriched RepU (ERU) is roughly two times larger than that after irradiation of the enriched natural uranium (ENU) because ^{236}U increases together with ^{235}U in the centrifuge re-enrichment process of RepU.

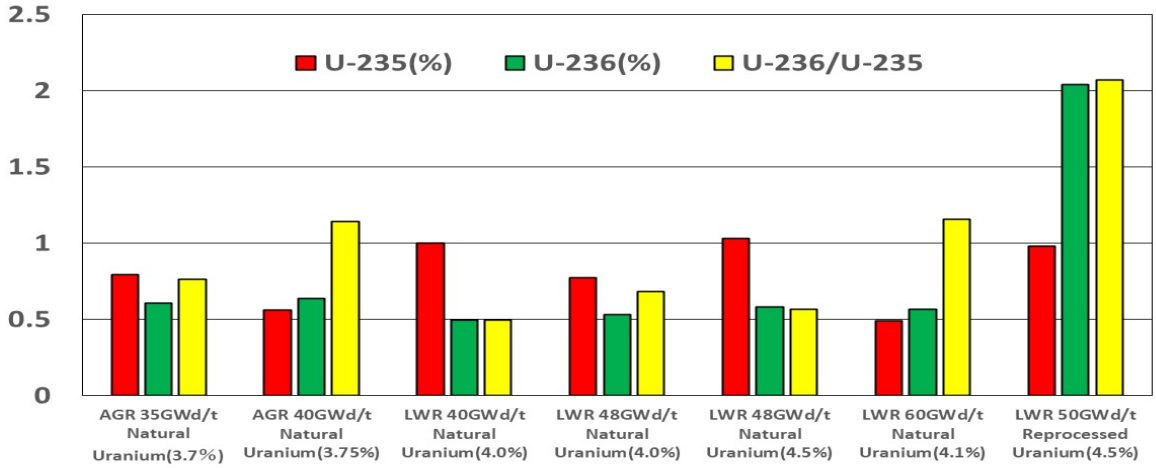

Figure 6. Examples of ^{235}U and ^{236}U after irradiation of UO_2 in AGR and LWR. (This figure is made by the authors using data from Ref. 20 and 21)

RE-ENRICHMENT OF REPROCESSED URANIUM

Currently, two technologies for U enrichment, centrifugation and gaseous diffusion, are in practical use. In the U enrichment cascade, all uranium isotopes, except ^{238}U, are enriched together with ^{235}U. The centrifuge process has several advantages compared to the diffusion process for the re-enrichment of RepU [20]. During the centrifuge re-enrichment of the remaining ^{235}U in the RepU, ^{236}U is also re-enriched together with ^{235}U. The isotopic ratios of ^{236}U to the remaining ^{235}U after re-enrichment of the RepU are roughly 0.3 ~ 0.4 for up to 5% ^{235}U enrichment process as shown in figure 7.

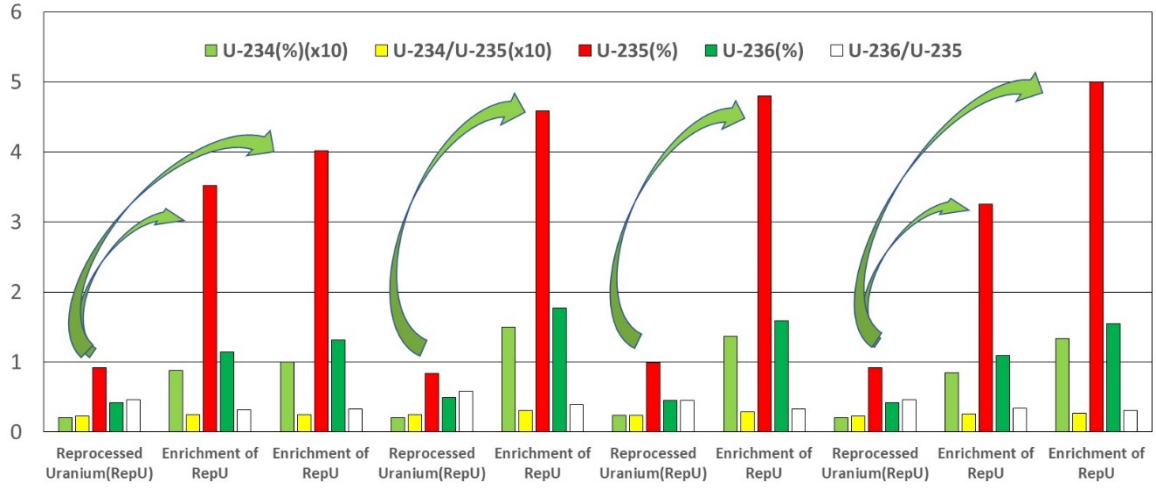

Figure 7. Illustration of re-enrichment of reprocessed uranium. (This figure is made by the authors using data from Ref. 20, 22, and 23)

$$\alpha_{236} = \frac{(U\text{-}236/U\text{-}235)_{Product}}{(U\text{-}236/U\text{-}235)_{Feed}} \quad (1)$$

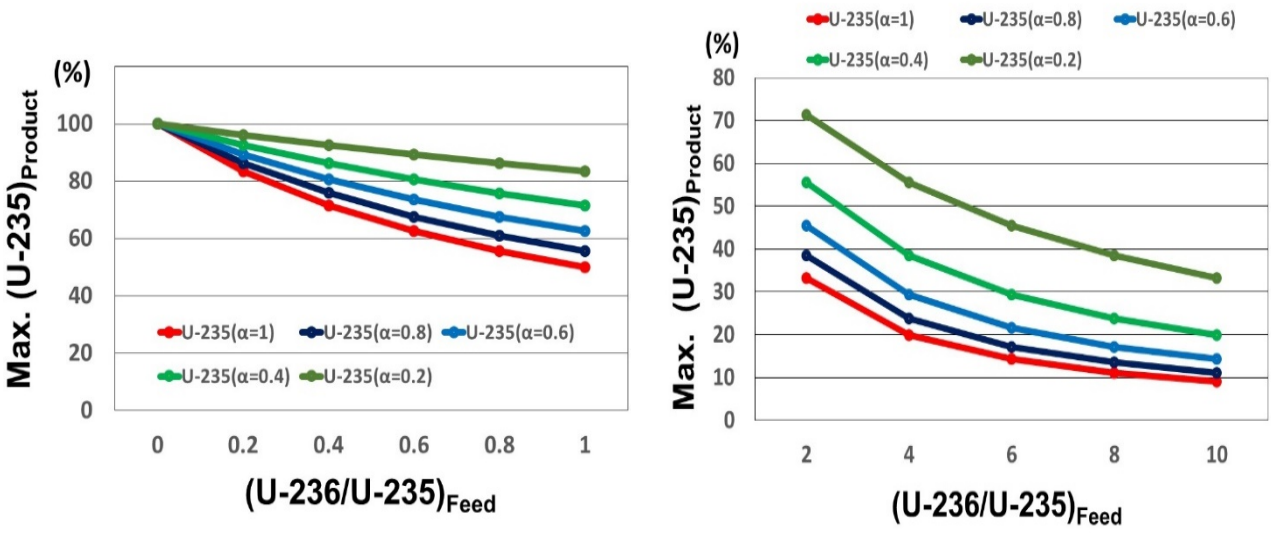

Figure 8. Attainable maximum possible enrichment of ^{235}U estimated by a simple model based on enrichment coefficient of ^{236}U (α_{236}) at ^{238}U=0%

To estimate the restriction of the re-enrichment (maximum enrichment) of ^{235}U due to the simultaneous enrichment of ^{236}U in the centrifuge enrichment of the RepU, a simple model based on the **Enrichment Coefficient of ^{236}U (α_{236})** of RepU defined in Eq. (1) can be utilized. When all of the ^{238}U is removed by the centrifuge process, with the exception of the ^{234}U isotope, the attained maximum enrichment of ^{235}U can be estimated by a simple model based on the Enrichment Coefficient of ^{236}U (α_{236}) of RepU, which are shown in figure 8.
Hence, the centrifuge re-enriching of the RepU makes it difficult to produce weapons-grade high-enriched ^{235}U because ^{236}U is also enriched together.

Based on the enrichment experimental data in papers [20,22,23], the Enrichment Coefficient of ^{236}U (α_{236}) were estimated, which are shown in figure 9. The average of the Enrichment Coefficients of ^{236}U (α_{236}) is about 0.7. The Enrichment Coefficients of ^{234}U (α_{234}) of RepU were also estimated, which is shown in figure 10. The average of the Enrichment Coefficients of ^{234}U (α_{234}) is about 1.16. As expected, since ^{234}U is lighter than ^{235}U compared to ^{236}U, Enrichment Coefficient of ^{234}U **(α_{234})** is greater than 1.

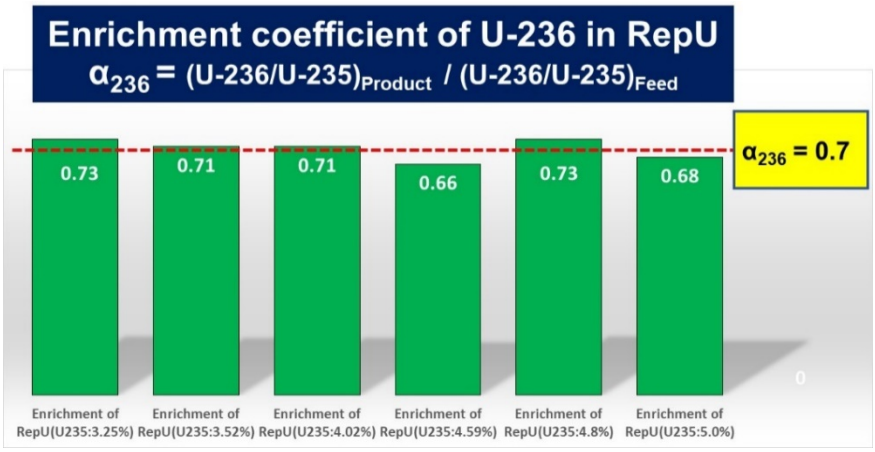

Figure 9. Enrichment Coefficients of ^{236}U (α_{236}) (This figure is made by the authors using data from Ref. 20, 22, and 23)

Based on figure 6, since the isotopic ratios of ^{236}U to ^{235}U in used fuel after a burnup of 40 GWd/t in AGR and 60 GWd/t in LWR are about 1.2, about

60% attained maximum enrichment of ^{235}U will be expected based on the data in figure 8. On the other hand, as shown in figure 6, since the isotopic ratio of ^{236}U to ^{235}U in used fuel after a burnup of 50 GWd/t of the re-enriched RepU in LWR is about 2, about 40% attained maximum enrichment of ^{235}U will be expected based on the data shown in figure 8.

After the re-enriched RepU is burned again in the reactor, naturally ^{235}U is reduced by fission to release energy during the burning and the content of ^{236}U is further increased compared to that after the first cycle. By repeating this process, the production of highly enriched weapons-grade U becomes more difficult from the multi-recycled RepU. The content of ^{238}Pu is also further increased compared to that after the first cycle, which means the PR of Pu as well as U is further increased after the first recycle.

PROLIFERATION RESISTANCE OF Pu AND U IN MULTI-RECYLCE OF REPROCESSED U

(1) Analysis of Reprocessed Uranium Multi-recycle

For a reactor physics lattice calculation, a first-collision probability method by SRAC (Ver.2002) code, which incorporates JENDL-3.3 with 107 group nuclear data library was used for the RepU multi-recycle analysis [21]. The fuel model conceived in this study was composed of three regions. A fuel and clad with outer radii 0.418 cm and 0.475 cm, respectively, with a light water moderator outside and a typical PWR fuel element pitch/diameter ratio of 1.38 was utilized.

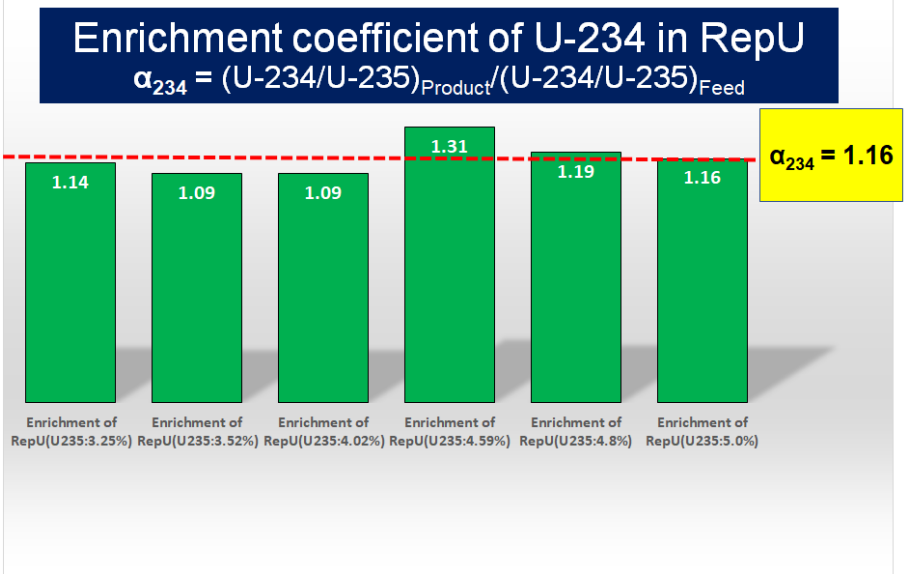

Figure 10. Enrichment Coefficients of ^{234}U (α_{234}) (This figure is made by the authors using data from Ref. 20, 22, and 23)

The uranium isotopic composition of the enriched RepU (ERU) was determined by a simplified ideal cascade calculation [24], which dealt with three major isotopes, ^{235}U, ^{236}U, and ^{238}U. The 5% ERU {^{235}U (5.0%), ^{236}U (1.61%), and ^{238}U (93.4%)} obtained by simplified cascade calculation of re-enrichment of 40 GWd/t burned used fuel after 5 years of cooling was used for the basic irradiation cycle. Envisaging recycling of the RepU fuel, a simple cascade calculation was repeated to reach 5% of ^{235}U in ERU by each re-enrichment process.

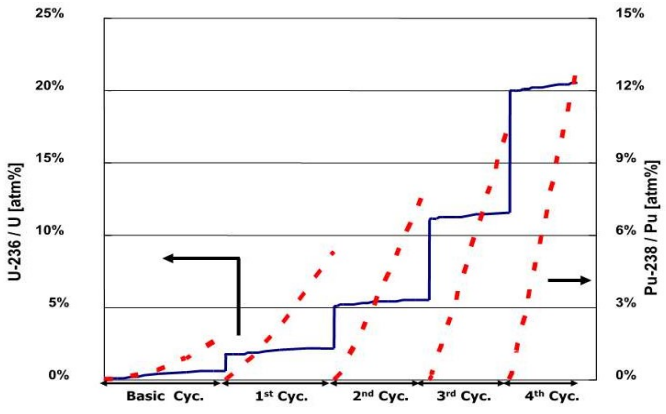

Figure 11. Changes in ^{236}U and ^{238}Pu isotopic compositions in RepU by repeating re-enrichment and irradiation [21]

A variation of ^{236}U concentration and ^{238}Pu build up in 5 % enriched natural uranium (ENU) during burnup for the basic fuel cycle and in 5 % ERU after the 1st cycle are calculated by SRAC (Ver.2002) code, the results of which are shown in figure 11.

From figure 11, one can notice that the ^{236}U concentration keeps increasing at each re-enrichment step in addition to a small increase during irradiation. Consequently, enhanced ^{236}U concentration entails the elevation of ^{238}Pu concentration in each cycle as well. At the end of cycle (EOC) of ENU irradiation in the basic cycle, build-up of ^{238}Pu is 1.9% in total Pu with 0.66% ^{236}U in total U, which is increased to 1.61% ^{236}U by centrifuge enrichment (ERU). The ^{238}Pu concentration upswings to 5.3% at the EOC of ERU 1st cycle due to the elevation of ^{236}U concentration in the ERU. So, the ^{238}Pu concentration in ERU increases step by step, ensuring the PR feature. Figure 12 shows a comparison of Pu isotopes' burnup behavior for the basic cycle of ENU and fourth cycle for the case of multi-recycle of RepU. The ^{238}Pu concentration in the fourth cycle is much higher than that in the basic cycle due to the elevation of ^{236}U.

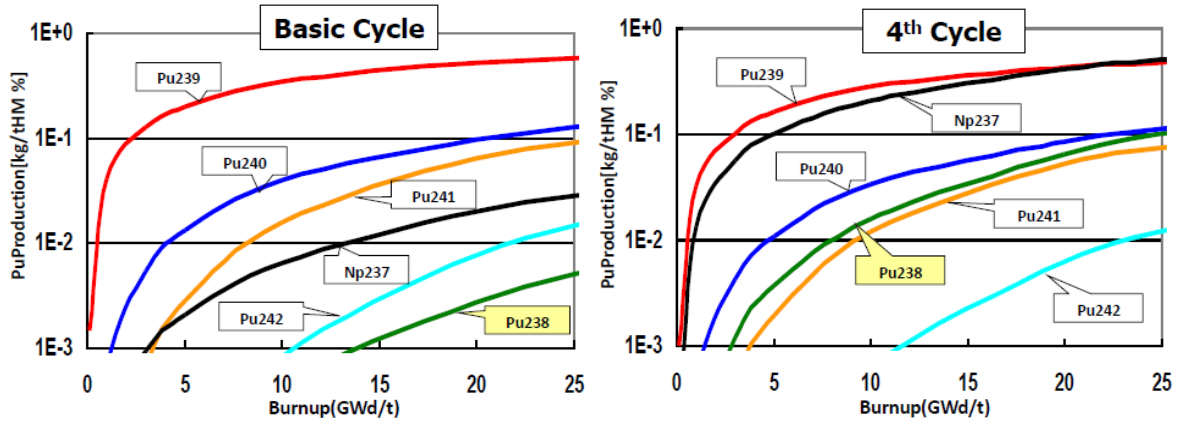

Figure 12. Comparison of Pu isotopes burnup behavior in basic cycle and in fourth cycle in multi-recycle of RepU [21].

(2) Enrichment Restriction of ^{235}U of Multi-recycle Reprocessed Uranium

As shown in figure 13, after the re-enriched RepU is burned again in the reactor, naturally ^{235}U is reduced by fission to release energy during the burning, and the content of ^{236}U after the second combustion is further increased compared to that after the first combustion. The ^{238}Pu content after the second combustion is also further increased compared to that after the first combustion,

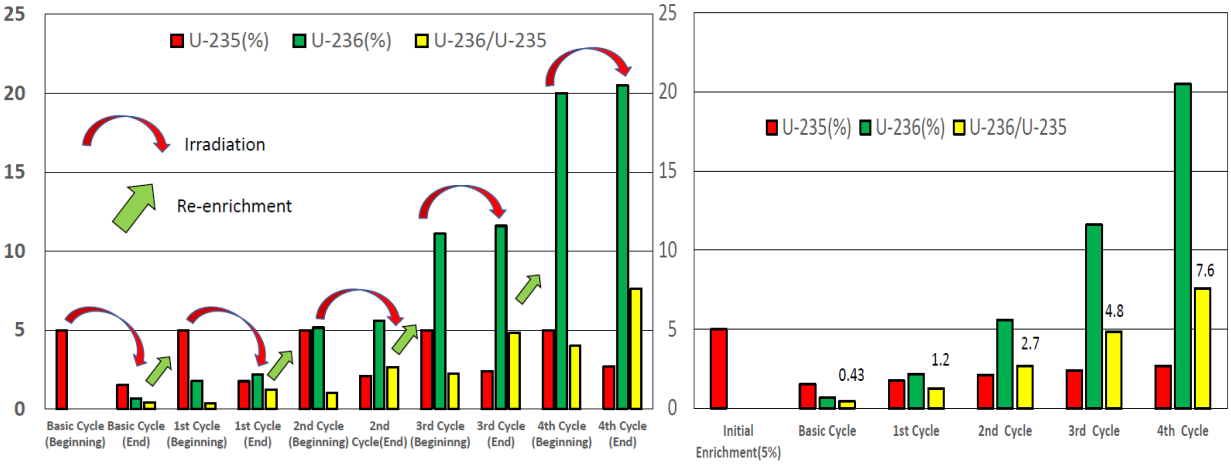

Figure 13. ^{235}U and ^{236}U after multi-recycle irradiations of RepU with 5% ^{235}U enrichment at each cycle.

which means the PR of Pu is further increased compared to that after the first combustion. By repeating this process, the production of highly-enriched weapons-grade U becomes more difficult from the multi-recycle reprocessed U.
The maximum enrichments of ^{235}U of RepU after burnup in each cycle were estimated by using a simple model with an enrichment coefficient of ^{236}U, $\alpha_{236} = 0.7$, and the results are shown in figure 14.

By the repeated recycle of RepU in LWR, the production of highly-enriched weapons-grade uranium becomes more and more difficult from the multi-recycled RepU. It is even difficult to produce enriched ^{235}U of more than 20%.

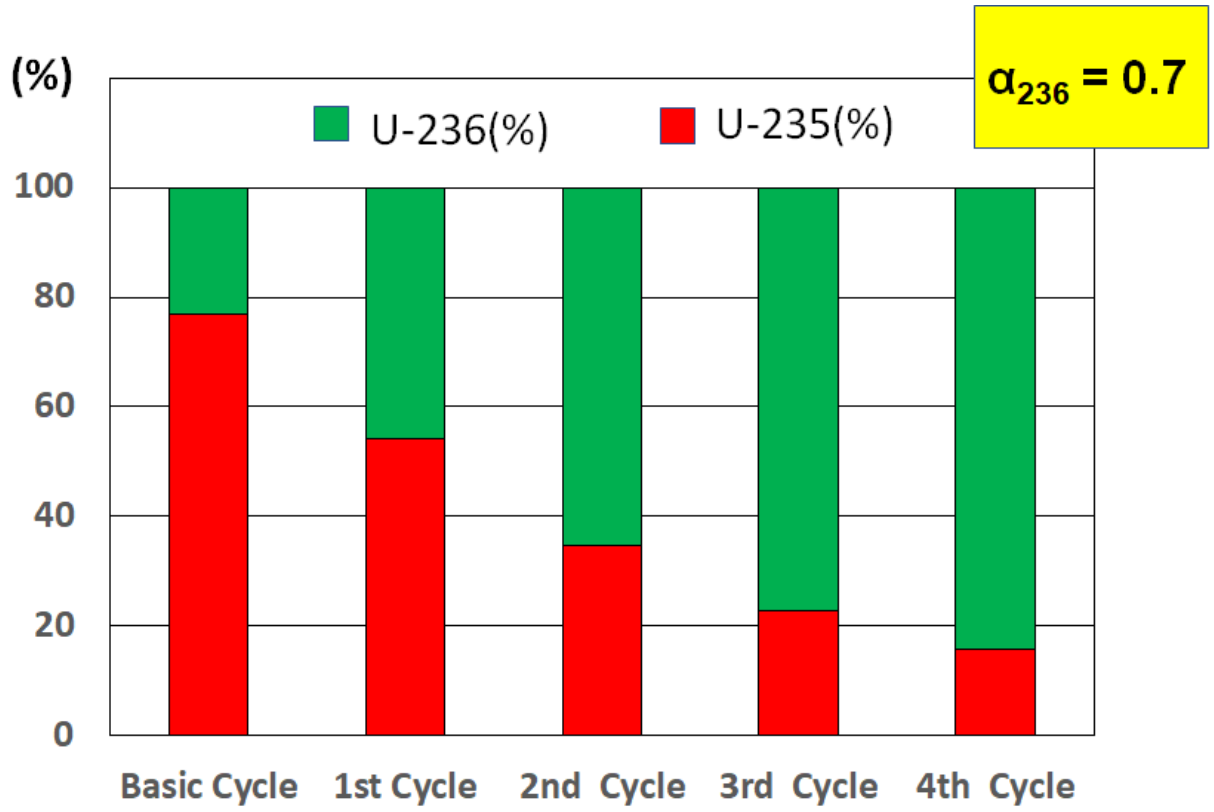

Figure 14. Attained maximum enrichment of ^{235}U estimated by using simple cascade model based on enrichment coefficient of ^{236}U ($\alpha_{236} = 0.7$) at $^{238}U = 0\%$.

The ^{236}U content in RepU is an attractive isotope to protect Pu and U from adversarial use. Recycling of minor actinides for the P^3 mechanism, proposed earlier by Saito, and the recycle of the RepU proposed here are both useful and important for the peaceful uses of nuclear energy in future.

CONCLUSIONS

The effects of multi-recycle of re-enriched RepU on the PR of Pu and U have been studied. The non-fissionable isotope ^{236}U is generated from ^{235}U during burnup in the nuclear reactor. During the burnup, by further absorbing neutrons, some of the ^{236}U content becomes ^{238}Pu via ^{237}Np/ ^{238}Np, which has strong PR characteristics because of its high decay heat and spontaneous fission neutron emission. During the centrifuge re-enrichment of ^{235}U in the RepU, ^{236}U is also re-enriched together with ^{235}U.

The general concept of enrichment coefficient (α_{236}) of ^{236}U, which is defined as the ratio of ^{236}U isotopic concentration to ^{235}U isotopic concentration after the enrichment compared to the ratio of ^{236}U isotopic concentration to ^{235}U isotopic concentration before the enrichment, is proposed.

Based on the centrifuge enrichment experimental data, the average of the enrichment coefficients of ^{236}U (α_{236}) is about 0.7.

After the re-enriched RepU is burned again in the reactor, naturally ^{235}U is reduced by fission to release energy during the burning, and the content of ^{236}U after that is further increased compared to that after the first cycle. The content of ^{238}Pu is also further increased compared to that after the first cycle, which means the PR of Pu is further increased compared to that after the first cycle. By repeating this process, the production of highly-enriched weapons-grade U becomes more difficult from the multi-recycled RepU. The ^{236}U content in RepU is an attractive isotope to protect Pu and U from adversarial use. The recycling of minor actinides for the P^3 mechanism, proposed earlier by Saito, and the recycle of RepU proposed here are both useful and important for the peaceful uses of nuclear energy and the move toward a "World without Nuclear Weapon."

The present theoretical prediction of the attained maximum enrichment of ^{235}U at ^{238}U= 0% by the simple model proposed here is recommended to be confirmed through experiments on the re-enrichment and irradiation of RepU.

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