Proceedings of the INMM & ESARDA Joint Annual Meeting
August 23-26 & August 30-September 1 2021, Virtual Meeting

Investigation of the Proliferation Resistance of Enriched Reprocessed Uranium Fuel Due to Higher Buildup of ^{238}Pu at Discharge

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ABSTRACT

Reprocessing of used nuclear fuel being the acquisition path for Pu by the proliferating states, the roles of intrinsic and extrinsic barriers to proliferation through this path need a deeper analysis. Denaturing Pu with higher fraction of ^{238}Pu is one of the intrinsic barriers that renders Pu less attractive for a nuclear explosive device (NED) and at the same time does not affect its value as a fuel for electricity generation. Previous studies suggest that the presence of ^{238}Pu fraction in excess of 6.2% in Pu provides high intrinsic proliferation resistance (PR). Hence, doping nuclear fuel with the precursors of ^{238}Pu, such as ^{237}Np had been recommended. A different precursor, ^{236}U, is a relatively easy isotope to incorporate in nuclear fuel and is the focus of this study. Enriched reprocessed uranium (ERU) contains higher ^{236}U (due to its presence in the used fuel and its further enhancement after ^{235}U re-enrichment) compared to natural or reprocessed uranium. This aspect can enhance intrinsic barriers against U and Pu proliferation while practicing the recycling of used fuel. Until this time, the verification of the PR of ERU has been conducted without a deeper understanding of the multi-isotope enrichment. ERU, containing multiple U isotopes, requires a different approach of enrichment calculation than the usual binary isotope method. Thus, to determine the concentration of ^{236}U in ERU, we used a matched-abundance ratio cascade (MARC) model, which has the advantage of being independent of a particular cascade type. We modified the MARC model to embody a gas centrifuge cascade, the most suitable commercial technique to handle ERU. Following the MARC model, Monte Carlo N-Particle Transport (MCNP) computer code was used to simulate fuel burnup of a pressurized water reactor (PWR) fuel assembly that uses ERU with an equivalent ^{235}U enrichment as that of a usual low enriched uranium (LEU) fuel assembly. Both, uranium enrichment and fuel burnup simulations allowed us to estimate the ^{238}Pu buildup in the ERU-based LWR fuel. As U recycling was repeated, the concentrations of ^{236}U and ^{238}Pu increased, and the sufficiently denatured Pu was obtained at the first U recycling. Several constraints regarding the utilization of ERU were also investigated. The maximum fuel burnup decreased leading to frequent refueling. The significant presence of ^{232}U and ^{234}U may lead to the radiological contamination of an enrichment facility. Moreover, the cascade should be re-configurated at each U recycling.

INTRODUCTION

Acquiring the needed fissile nuclear material is a difficult step in building a nuclear explosive device (NED). Thus, it is important to raise the technical barriers to deter the states from acquiring fissile nuclear material. In 1978, Oak Ridge National Laboratory (ORNL) scientists introduced an idea of denaturing plutonium (Pu) with the ^{238}Pu isotope to hinder the production of WgPu [1]. DeVolpi defined such denaturing operation as rendering a fissile nuclear material less suitable for producing a NED [2]. When Pu contains a high concentration of ^{238}Pu, the material becomes unfavorable in terms

of heat-generation rate, spontaneous neutron generation rate, and other radiation emission rate. The ^{238}Pu level can be artificially boosted by introducing ^{238}Pu precursors such as ^{236}U, ^{237}Np, ^{241}Am, and ^{242}Cm into the fuel. Among these precursors, this study focused on the contribution of ^{236}U to the proliferation resistance (PR) of Pu. Separating ^{236}U from the fuel using U enrichment technology is challenging because the atomic mass of ^{236}U is different from that of ^{235}U only by one unit. A number of studies have been conducted focusing on the removal of ^{236}U from the fuel, which demands a complex U separation process [3]. ^{236}U is a neutron poison [4]. Nevertheless, ^{236}U is a suitable isotope to increase the PR of the fuel cycle. When enriched natural uranium (ENU) is irradiated in a reactor, the probability of ^{235}U absorbing a neutron and not resulting in a fission but producing ^{236}U is 18%. Even though some of the ^{236}U get converted to ^{238}Pu, some of the ^{236}U will remain in the used fuel, which will be found in reprocessed U (RepU). When enriched RepU (ERU) is produced by re-enriching RepU, the concentration of ^{236}U is further enhanced, which will increase the PR of ERU (because further ^{235}U enrichment to weapons grade U will enrich ^{236}U simultaneously) as well as that of the irradiated ERU (because of the enhanced production of ^{238}Pu from ^{236}U).

The objective of this study was to analyze the PR of ERU and irradiated ERU in a reactor by estimating the amount ^{236}U in ERU and the amount of ^{238}Pu thus produced in irradiated ERU recycled multiple times in a light water reactor (LWR). Multicomponent uranium isotope enrichment calculation followed by neutronics simulation of fuel burnup were the methods used for this study. Previous studies on ERU's potential of producing denatured Pu overlooked the multicomponent uranium enrichment calculation [5, 6]. Moreover, the studies that dealt with multicomponent uranium enrichment calculation did not survey the PR of ERU [7, 8]. The matched-abundance ratio cascade (MARC) model was utilized for multicomponent uranium enrichment calculations and Monte Carlo N-Particle transport code (MCNP) was utilized for fuel burnup simulations.

METHODOLOGY

The multi-recycling scenario of used fuel consists of five steps: (1) simulation of natural U (NatU) enrichment to 5 wt% ^{235}U by a gas centrifuge cascade is carried out using MARC model to produce ENU; (2) Irradiation simulation of the fuel assembly with ENU fuel is carried out using MCNP code; (3) The irradiated (used) fuel is then cooled for five years and reprocessed without considering material loss in the process; (4) The RepU is re-enriched to a ^{235}U enrichment of 5 wt% using MARC model; and (5) Steps 2 to 4 were repeated five times with ERU to simulate multiple recycling in an LWR. It was assumed that there is no loss of materials during the recycling process and no decay of materials in between the aforementioned steps.

Modeling and Simulation of the Enrichment of Reprocessed Uranium

Introduced by de la Garza, the MARC is a multicomponent U enrichment cascade model that is suitable to model and simulate the enrichment of RepU containing multiple U isotopes of U [9 - 11]. The MARC model is a theoretical cascade that has the minimum number of separating elements and hence the most cost-effective. It should be noted that RepU has higher initial ^{235}U concentration than NatU. As a result, the common tails enrichment, which is from 0.25 to 0.30 wt% cannot be applied in the case ERU. Instead, the waste optimization should be carried out using

$$C_P = C_F f_{F/P}(x_W) + C_U f_{SWU/P}(x_W), \quad (1)$$

where C_P, C_F, and C_U are the total cost per product, the cost per feed, the cost per separative work unit (SWU) respectively; $f_{F/P}(x_W)$ is the feed per product as a function of tails enrichment; $f_{SWU/P}(x_W)$ is the separative work per product as a function of tails enrichment [12]. The feed per product can be expressed as

$$f_{F/P}(x_W) = \frac{x_P - x_W}{x_F - x_W}, \tag{2}$$

where x_P, x_W, and x_F are isotopic composition of any component in the product, waste, and feed streams, respectively. The SWU per product, however, involves a complicated value function expressed as

$$f_{SWU/P}(x_W) = PV_{i,P} + WV_{i,W} - FV_{i,F}, \qquad i = 1,2,\cdots,m, \tag{3}$$

$$V_i = \frac{(\sqrt{q_j} - 1)}{(\sqrt{q_j} + 1)} \ln \sqrt{q_j} \sum_{i=1}^{m} \frac{x_i \ln \frac{x_j}{x_k}}{\frac{(q_i - \sqrt{q_j})}{(q_i + \sqrt{q_j})} \ln \sqrt{q_j}}, \tag{4}$$

where P, W, and F are the flow rate of the product, waste, and feed streams, respectively; V_i is the value function of the i^{th} component; j and k are the components of which the relative abundance matches at the mixing points; and q_i is the overall stage separation factor of the i^{th} component. In addition, the two components, j^{th} and k^{th} components, that meets the matched-abundance condition must be determined. The matched-abundance components affect the total flow rate of a cascade, or the total number of separating elements in a cascade. The total flow rate equations (Eqs. 5 & 6) are analogous to the SWU per product equation. The minimum total flow rate is achieved when ^{235}U and ^{238}U are the matched-abundance components regardless of head or tails enrichment [13].

$$L_{total} = Pl_P + Wl_W - Fl_F, \qquad i = 1,2,\cdots,m, \tag{5}$$

$$l = \sum_{i=1}^{m} \frac{x_i \ln \frac{x_j}{x_k}}{\frac{(q_i - \sqrt{q_j})}{(q_i + \sqrt{q_j})} \ln \sqrt{q_j}}. \tag{6}$$

Lastly, the MARC model is modified to embody a gas centrifuge cascade because ERU is produced only with gas centrifuge enrichment technology due to its technical advantages [14, 15]. The stage separation factor used in our MARC model was modified in accordance with its separation method suggested by H. Wood [16].

Simulation of Fuel Burnup and Cooling in an LWR

A Westinghouse AP1000 LWR fuel assembly model was used to simulate the burnup of ENU and ERU fuels using MCNP [17 - 19]. There were no neutron absorbing rods in the fuel assembly and the coolant was borated water. Fuel burnup simulations were carried out for each cycle until the effective multiplication factor (k_{eff}) reached unity. The discharge burnup of ENU was 42 GWd/MTU. After the burnup and cooling, the RepU extracted from the used fuel was re-enriched to produce an ERU fuel.

Proliferation Resistance Analysis of Plutonium

This study utilized the criterion proposed by Y. Kimura et al. to determine whether denaturing of Pu can be achieved or not [20]. In his study, a hypothetical nuclear explosive device (HNED) made from reactor-grade Pu (RGPu) was modeled and investigated how much ^{238}Pu would limit the production of HNED due to the decay heat. Additionally, Kimura went a step further and included the contribution of ^{240}Pu and ^{242}Pu to the decay heat. The scientific limit for an HNED is listed in Table 1. The technology class indicates the capability of the state to handle the decay heat while manufacturing an HNED.

Table 1. Scientific limit for hypothetical nuclear explosive device [20]

Technology class	Limit of sum of even-mass number Pu isotopes
Low	1.9 wt% = ^{238}Pu (wt%) + 0.02 × [^{240}Pu (wt%) + ^{242}Pu (wt%)]
Medium	6.2 wt% = ^{238}Pu (wt%) + 0.05 × [^{240}Pu (wt%) + ^{242}Pu (wt%)]
High	15.0 wt% = ^{238}Pu (wt%) + 0.11 × [^{240}Pu (wt%) + ^{242}Pu (wt%)]

RESULTS AND DISCUSSION

The Effect of ^{236}U on the Fuel Cycle

For every U recycle, ERU fuels are re-enriched to the same 5 wt% ^{235}U. As U recycling was repeated, the relative abundance of ^{236}U to ^{235}U increased and the maximum fuel burnup decreased from 42 to 22.3 GWd/MTU due to the negative neutron poison effect of accumulated ^{236}U. The ^{236}U/^{235}U ratio for each cycle before and after U enrichment is shown in Fig. 1. The residual ^{235}U increased in the used fuel after each fuel cycle. On average, number of used fuel assemblies required from the preceding U cycle to generate the same amount of power as ENU cycle was 4.43.

Denatured Plutonium

The Pu compositions estimated in the used fuel for each cycle are reported in Table 2. The degree of Pu denaturing is also reported in Table 2. From Table 2, it can be inferred that the ^{238}Pu buildup was enhanced by the presence of ^{236}U. Denatured Pu for low technology class was instantly achieved with ENU. Denatured Pu for medium technology class was achieved at the first U recycling. Meanwhile, it was feasible to produce denatured Pu for high technology class only after the fifth U recycling. It is still an encouraging result that denatured Pu for the low and medium technology classes can be attained after first recycling. The states falling under the high technology class already have the experience with NED manufacturing. Therefore, building an NED from RGPu would not be the choice of those high technology states.

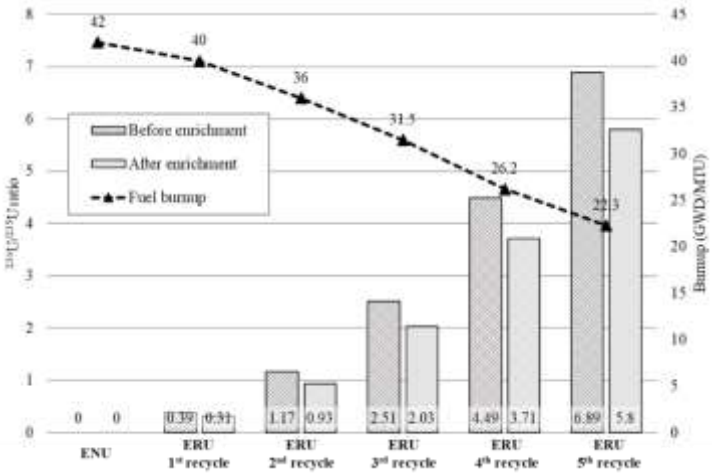

Figure 1. Relative abundance of ^{236}U to ^{235}U and the corresponding maximum fuel burnup.

Table 2. Plutonium weight fraction in used fuel and denatured Pu comparison for each cycle.

Cycle	^{238}Pu (wt%)	^{239}Pu (wt%)	^{240}Pu (wt%)	^{241}Pu (wt%)	^{242}Pu (wt%)	^{238}Pu (wt%) + z × [^{240}Pu (wt%) + ^{242}Pu (wt%)]		
						Low Technology (z=0.02)	Medium Technology (z=0.05)	High Technology (z=0.11)
ENU	1.81	60.18	22.26	11.14	4.60	2.35	3.16	4.77
ERU 1st recycle	5.13	59.61	20.83	10.47	3.96	5.63	6.37	7.86
ERU 2nd recycle	7.82	60.46	19.02	9.59	3.11	8.26	8.93	10.25
ERU 3rd recycle	10.23	61.72	17.14	8.60	2.31	10.62	11.20	12.37
ERU 4th recycle	12.08	63.78	15.12	7.44	1.57	12.41	12.91	13.92
ERU 5th recycle	14.02	65.03	13.42	6.43	1.11	14.31	14.75	15.62
Scientific limit for failure of hypothetical nuclear explosive device (HNED)						**1.90**	**6.2**	**15.0**

Compensation for Loss in Fuel Burnup in ERU

As shown earlier in Fig. 1, the maximum possible fuel burnup kept on decreasing which would lead to frequent refueling. In order to compensate for the neutron poison effect of ^{236}U, ERU must be enriched with excess amount of ^{235}U than the 5 wt%. However, the excess amount of ^{235}U would bring about the ^{235}U enrichment above the limit established by the American Society for Testing and Materials (ASTM) and the US Nuclear Regulatory Commission (NRC) [21 - 23]. Even if the U were enriched above the limit, it would eventually become unfeasible to reach equivalent k_{eff} value after several repetitive recycling due to the increasing concentration of ^{236}U.

Besides the ^{236}U neutron poison effect, the radiological hazards at the U enrichment facility will increase after each recycle. This is because of the buildup of the daughter products of ^{232}U and ^{234}U that emit high-energy gamma radiation. Thus, the ASTM regulates the concentration of ^{232}U and ^{234}U at the enrichment facility below 5E-7 wt% and 0.048 wt%, respectively. The buildup of ^{232}U and ^{234}U for each cycle are shown in Figs. 2 and 3, respectively. The concentration of ^{234}U exceeded the ASTM limit at

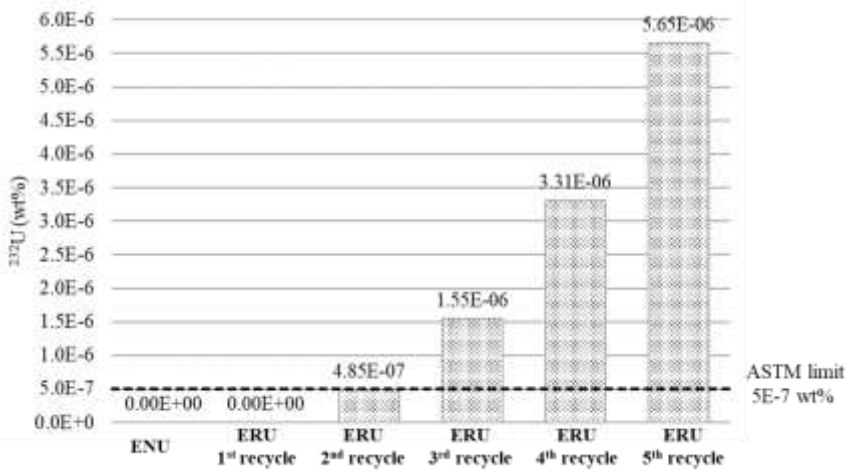

Figure 2. ^{232}U inventory of U product.

the initial U recycling and the concentration of ^{232}U at the third U recycling. It may be noted that the concentrations of ^{232}U and ^{234}U were enhanced due to U enrichment.

It should be pointed out that the composition of RepU will vary depending on the fuel burnup, type of reactor, reactor power, cooling period, etc., and this study analyzed the fuel recycling for a specific reactor at a given reactor power and cooling period. The U enrichment cascade must be fine-tuned according to the feed composition. Otherwise, it will be less cost-effective, and the head enrichment will fall short of expectations. The difference feed composition at each recycling will bring about an additional burden to the U enrichment facility.

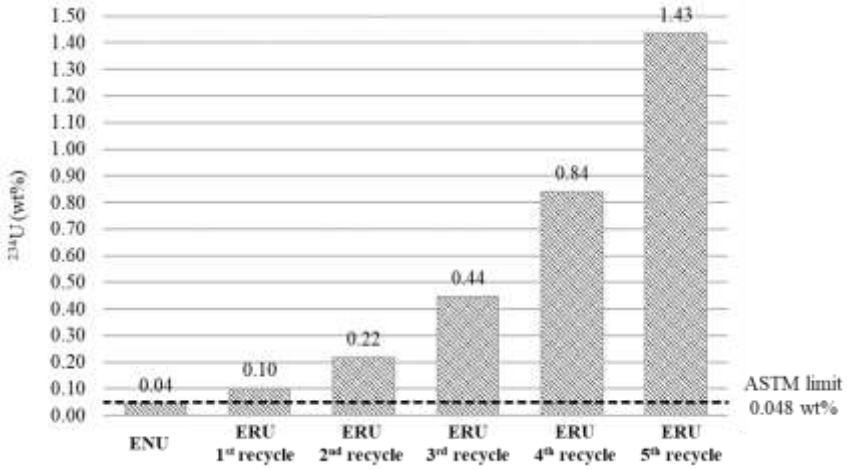

Figure 3. ^{234}U inventory of U product.

CONCLUSION

The inherent intrinsic proliferation resistance (PR) of enriched reprocessed uranium (ERU) with respect to the production of a plutonium-based nuclear explosive (NED) was the focus of this study. Results of this study showed that the employment of U recycling resulted in the efficient use of U resources with high proliferation resistance and could support the reduction of the volume of the deep geological repository. According to Kimura et al., [19], Pu containing more than 6.2 wt% ^{238}Pu can be considered denatured due to its high decay heat. In order to verify the viability of producing denatured Pu with ERU, two consecutive calculations were carried out. First, a modified matched-abundance ratio cascade (MARC) model was used to simulate the enrichment of U with multicomponent (multi isotopes of U). Next, a single pressurized water reactor (PWR) fuel assembly is modeled using Monte Carlo N-Particle code to simulate the fuel burnup and cooling of low enriched uranium (LEU). Denaturing of Pu for low and medium technology classes of states was found to be attained at the first U recycling period. As recycling is repeated, the value of ERU as a source of power continue to degrade due to the increasing concentration of U isotopes, specifically ^{236}U. Based on the results, it is worth applying ERU to produce denatured Pu, but U recycling will have challenges due to other constrains such as the need for higher ^{235}U enrichment than 5 wt% and radiation hazards associated with the decay of minor U isotopes (^{232} and ^{234}U).

ACKNOWLEDGEMENT

The Republic of Korea Army funded the graduate study of one of the authors, Saehyun Choi to conduct this study at Texas A&M University.

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