PROLIFERATION RESISTANCE ANALYSIS OF MULTIPLE RECYCLING OF RE-ENRICHED REPROCESSED URANIUM FUEL IN COMMERCIAL LIGHT WATER REACTORS

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ABSTRACT

The rapid growing demand for electricity combined with the limitations of renewable energy such as wind and solar and also excessive CO_2 emission from fossil fuels make nuclear power more attractive as an important supply of energy. However, to develop a robust nuclear regime, one of the aspects that should be considered is nuclear non-proliferation. The sensitive nuclear proliferation parts of nuclear fuel cycles such as reprocessing of spent fuel has been subject of many studies over years. Proliferation resistance assessment of re-enriched reprocessed uranium fuel discharged from a light water reactor water-cooled water-moderated energetic reactor (VVER) is analyzed in this study by estimating the growth in the desired amount of even numbered uranium and plutonium isotopes, especially ^{236}U and ^{238}Pu to make the spent fuel less attainable for military purposes. During the fuel burnup non-fissile uranium isotope, ^{236}U is considered for proliferation resistance as one of the signature isotopes for the reprocessed uranium (RepU). ^{236}U is generated from its precursor ^{235}U by successive neutron capture, and one of the reaction chains to produce ^{238}Pu is transmuting minor actinides starting from ^{237}Np, which is a result of beta decay of ^{236}U. Multiple recycling enables ^{236}U concentration to keep growing after each centrifuge re-enrichment and irradiation thus resulting in contributing to ^{238}Pu development in the spent fuel. ^{238}Pu denatures Pu because of its high spontaneous fission neutron emission rate and decay heat. Also, the spent fuel at the end of multiple recycling accumulates ^{236}U in the fuel in such a way that further re-enrichment of ^{235}U does denature U due to the co-enrichment of ^{236}U with ^{235}U.

INTRODUCTION

Nuclear energy has been attracting attention for energy supply, but it is also appealing for military or nonpeaceful purposes. The accumulation of secondary nuclear materials such as Pu, minor actinides, and RepU present potential risk with respect to military purposes. According to

nuclear weapon states, a nuclear weapon could be built with plutonium having practically any mix of isotopes since all of them are fissionable. However, the odd isotopes (^{239}Pu and ^{241}Pu), both fissile meaning that fission could occur in any neutron energy slow or fast, have an advantage of requiring less bare critical mass [1]. For weapon applications, plutonium containing very high percentages of the isotope ^{239}Pu (relatively pure) is much preferable because it is formed from ^{238}U by neutron capture, and it is separated after a brief radiation (low burnup) which is inefficient for power production. However, even plutonium from high burnup commercial reactors (less than 90% of ^{239}Pu) may be utilized to create nuclear bombs with significant explosive yields since the plutonium in the spent fuel taken from a commercial light water reactor (LWR) (burn-up of 42 GWd/t) is around 53% of ^{239}Pu.

In discussion for the technical assessment of preventing proliferation, there are two terms that are usually used: non-proliferation and proliferation resistance (PR). Non-proliferation refers to preventing the spread and the increase of nuclear weapons. In order to achieve and serve this goal, the International Atomic Energy Agency (IAEA) was established, and numerous weapons control and non-proliferation treaties have been put into effect by this global authority. The definition of PR is relatively well-agreed upon as: The characteristics of a nuclear energy system that impede the diversion of undeclared production of nuclear material or misuse of technology by states in order to acquire nuclear weapons or other nuclear explosive devices [2]. While there are regulatory measures such as the IAEA nuclear safeguards, which gives a high degree of assurance of compliance with authority through international verification, strengthen the political commitments of States against nuclear weapons, it is also significantly important to have a non-proliferation aspect to put forward by experts with the aim of developing new technologies to increase PR because there is no such thing as a proliferation-proof nuclear fuel cycle.

Given these circumstances, there might be intrinsic features of nuclear fuel and extrinsic measures that could impede the development of nuclear explosive devices. For intrinsic features, most spent fuel from power reactors contains even mass numbered isotopes levels that make it less efficient for use in a nuclear explosive. The effectiveness of these intrinsic features might not be enough itself and need to be supported by extrinsic measures. This paper presents some of the intrinsic features with the support of different technologies. To evaluate the suitability of an isotope for an explosive device, the proliferation metrics (isotopic barrier effectiveness) could be used. The metrics consist of some quantities for constructing a nuclear weapon and they are the amount of nuclear material required to build a weapon (critical mass), the heat generation rate, and spontaneous fission rate. Critical mass is vital because large critical mass leads to a heavy and large weapon which makes it difficult to deliver the weapon. Heat generation rate is another point to be evaluated due to the fact that it requires a stronger barrier to keep the explosive components from melting. Lastly, the spontaneous neutron emission rate of the material has a huge impact on designing nuclear weapons because excessive neutron emission may cause the nuclear explosive device to ‘preinitiate’ which makes the weapon useless [3]. For ^{238}Pu, the decay heat from alpha emission and the spontaneous neutron emission rate are significantly high compared to other isotopes which makes this isotope less attractive. There are different ways to accumulate ^{238}Pu. Previous studies suggested that one way to accumulate ^{238}Pu could be by doping nuclear fuel with the minor actinides which are the precursor of ^{238}Pu, such as ^{237}Np or

^{241}Am thus increasing the isotropic barrier of the spent fuel [4], second way could be multiple recycling of nuclear spent fuel without adding minor actinides thus denature the plutonium, which is the main focus of this thesis.

METHODOLOGY

The nuclear power reactor model chosen for this study is a VVER-1000 MWe fuel assembly. After choosing the reactor type, the fuel assembly is simulated for fuel burnup by carrying out neutron transport using MCNP6.2 code [5]. After gathering all the information for the geometry of the VVER, MCNP models of fuel assemblies were prepared for neutronics assessment using the KCODE and BURN feature of MCNP6.2. Following the fuel burnup simulations including one year of cooling, the spent fuel is assumed to be reprocessed to obtain the RepU to be re-enriched. The code used for re-enriching the spent fuel is MARC, which stands for Matched Abundance Ratio Cascade model [6]. To what degree it is enriched in ^{235}U, depends on the effective neutron multiplication factor, k-eff at the reactor starting point. The isotopes fractions were read out of the MCNP6.2 code output to determine the buildup of U and Pu isotopes separately especially U^{236} and Pu^{238} to carry out the the PR assessment. The RepU is re-enriched to 4.35wt% ^{235}U and the buildup of ^{238}Pu is estimated in the fuel assembly by recycling the re-enriched RepU, which can denature plutonium.

SIMULATION OF VVER

Russian VVER-1000 fuel assembly model was used to simulate the burnup of enriched natural uranium (ENU) and re-enriched reprocessed uranium (ERU) fuels using MCNP6.2. The model is a typical VVER-1000 fuel assembly lattice with 312 fuel pins with 18 control rod guide tube locations and 1 central instrumentation channel [7] (ref. Figure 1). The assembly for this research has two different ^{235}U enrichment 3.3wt % and 3.7wt %. There is no neutron absorber in fuel rods, and coolant is light water. For each cycle, fuel burnup simulations are carried out until the infinitive multiplication factor (k-inf) reaches unity and potentially the fuel burnup is 33 GWd/MTU k-inf=1.

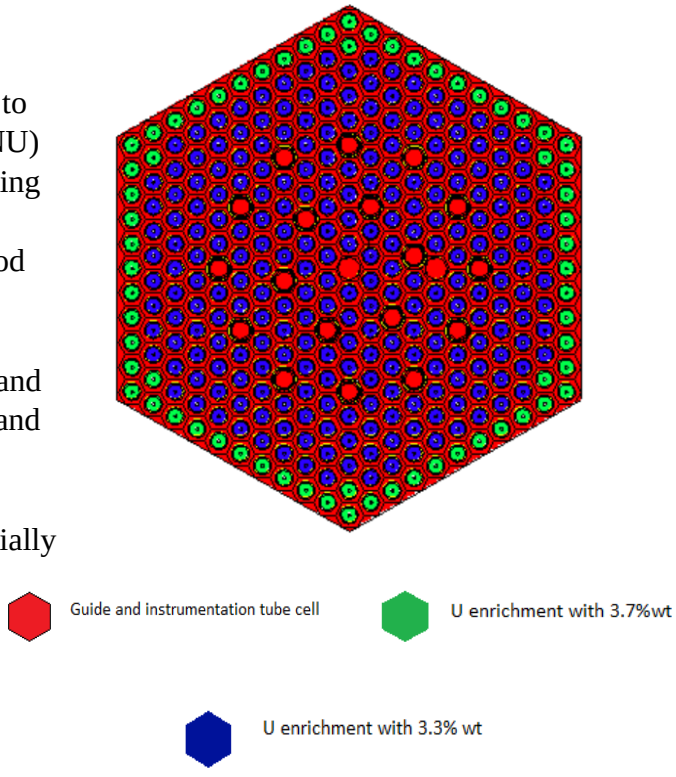

Figure1. Axial Cross-sections of a Single Fuel Assembly

RESULTS AND DISCUSSIONS

Reprocessed Uranium from VVER

Natural uranium only contains 0.72 wt% of ^{235}U, 0.0054 wt% of ^{234}U and 99.275 wt% of ^{238}U [8] and as indicated before, the standard limit of enrichment for a commercial LWR varies from 3 to 5 wt%. During the fuel burnup, in addition to these three isotopes, ^{236}U, considered as one of the signature isotopes for the RepU, is produced by the neutron capture of ^{235}U that does not lead to a fission. ^{236}U does not pose an external radiological hazard because it is a long-lived alpha emitter. However, it absorbs a neutron to create ^{237}U, which then beta decays to ^{237}Np. This process removes neutrons to a significant extent from the chain reaction which is depicted in the chain reaction scheme in Figure 2 [9]. Due to the neutron absorption of ^{236}U present in RepU, higher than the usual LWR fuel ^{235}U enrichment is needed to compensate for the negative reactivity so that the reactor cycle length is comparable to the ENU assemblies. In this study the initial ^{235}U enrichments considered are 3.7 wt% and 3.3 wt%, which are in the same in the same fuel assembly, because the peripheral pins have a different enrichment. The enrichment for the RepU was decided as 4.5wt% considering the reactivity loss due to the enhanced presence of ^{236}U compared to ENU. Figure 3 below illustrates ^{235}U and ^{236}U masses in the fuel assembly after 33 GWd/t burnup and for the cases of re-enrichment of uranium for the multiple recycling.

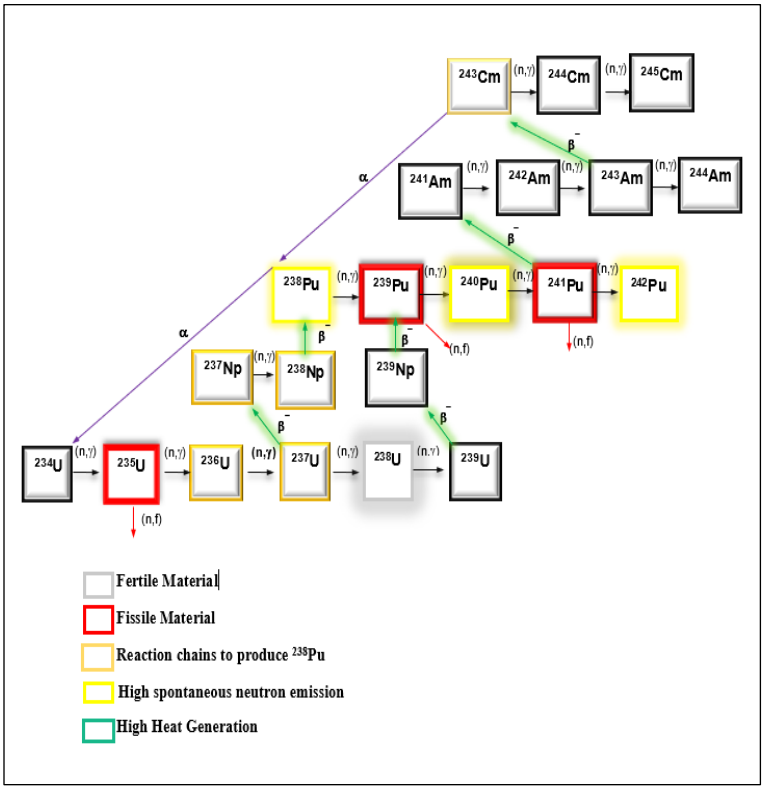

Figure 2. Principal chains of nuclear reactions leading to **^{238}Pu.**

During re-enrichment of remaining uranium by gas centrifuge, the concentration of ^{236}U also goes up due to the co-enrichment with ^{235}U. As listed in Table 1 and Figure 3, the build-up of ^{236}U at the end of cycle (EOC) of ENU irradiation is 0.47 wt% in total U, which increased to 1.40 wt% ^{236}U by centrifuge enrichment (ERU), indicating PR will be even further enhanced with recycle because of the enhanced ^{238}Pu build up from ^{236}U. These findings imply that the ERU fuel becomes more resistant to proliferation following the second cycle of irradiation and re-enrichment thus makes highly enriched weapon grade uranium harder to produce from multiple recycling.

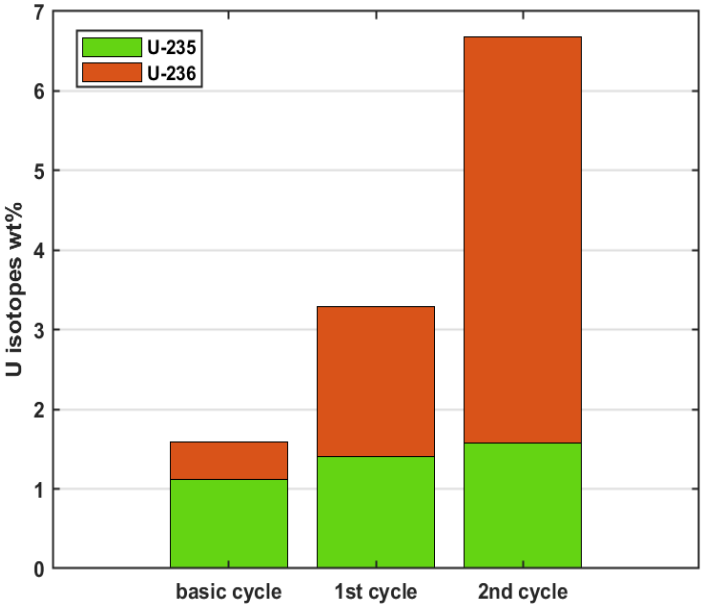

Figure3. Changes in ^{236}U composition in RepU.

ISOTOPES	^{234}U (%)	^{235}U (%)	^{236}U (%)	^{238}U (%)
(Basic cyle)	3.61E-3	1.12	0.47	98.4
Product	1.67E-2	4.35	1.40	94.2
RepU (1_{st} cyle)	1.17E-2	1.41	1.88	96.7
Product	4.01E-2	4.35	4.65	91.0
RepU (2_{nd} cyle)	2.89E-2	1.58	5.10	93.3

Table1. Isotropic concentration of U isotopes after each reprocessing and re-enrichment.

Protected Plutonium Production of Recycling of RepU

Denaturing Pu could be characterized by the isotopic barrier effectiveness, spontaneous fission, and heat generation rate, which could be enhanced by increasing the fraction of even number of plutonium isotopes, especially ^{238}Pu and ^{240}Pu. Although recent studies to date have utilized ^{238}Pu to a great extent, ^{240}Pu is another isotope that should be considered in the case of PR owing to its high spontaneous fission rate. ^{240}Pu has the second highest emission of spontaneous fission

rate among Pu isotopes and its fraction in total Pu is higher than ^{238}Pu in a regularly discharged spent fuel, which enables this isotope as one of the key factors in determining the PR.

One of the reaction chains to produce ^{238}Pu is transmuting minor actinides starting from ^{237}Np which is a result of beta decay of ^{237}U, which is produced by the neutron capture of ^{236}U and ^{240}Pu production is a result of neutron capture of ^{239}Pu. According to the IAEA, plutonium with an isotopic concentration of ^{238}Pu exceeding 80% is referred as denatured or proliferation resistant and could be exempted from safeguards and it would be treated by the IAEA equivalent to low enriched uranium (LEU) [10]. The question is, would ^{238}Pu content significantly lower than 80 wt% be sufficient to denature Pu to be used in nuclear explosive devices? It was found that, even for high technology nuclear explosives, ^{238}Pu content higher than 6-8 wt% would not be effective to use in nuclear explosive devices [11].Figure 4 shows how multiple recycling of RepU enables the accumulation of ^{238}Pu to enhance the protected plutonium production. As mentioned earlier, ^{236}U concentration keeps growing after each re-enrichment and irradiation thus resulting in contributing to ^{238}Pu buildup in the spent fuel. At the end of cycle of ENU, buildup of ^{238}Pu is around 1.69 wt% from 33 GWd/t burnup and the concentration jumps to 8.07 wt% at the EOC of 3rd recycle as a result of ^{236}U build up EOC of 2nd recycle.

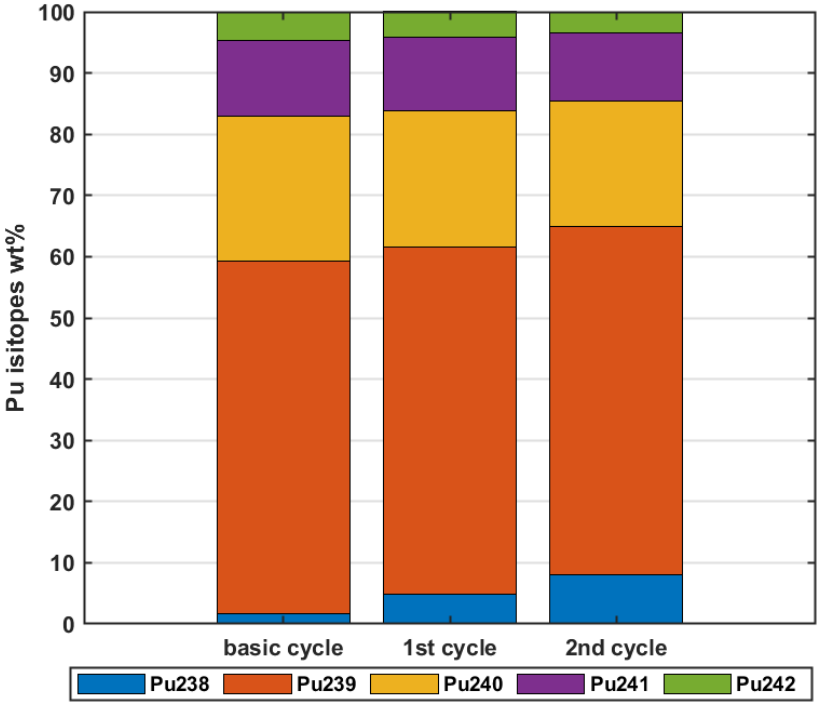

Figure 4. Changes in **^{238}Pu** compositions in RepU.

Isotopes (%)	^{239}PU	^{239}PU	^{240}PU	^{241}PU	^{242}PU
Basic cyle	1.69	57.5	23.8	12.4	4.61
RepU (1$_{st}$ cyle)	4.90	56.7	22.2	12.0	4.21
RepU (2$_{nd}$ cyle)	8.07	56.8	20.5	11.1	3.46

Table2. Isotropic concentration of Pu isotopes after each reprocessing.

CONCLUSION

This study focused on the feasibility of ERU in Russian design LWR (VVER) with respect to military and non peacefull purposes. The results of this study demonstrated that using reprocessed re-enriched uranium resulted in the recovery of uranium isotopes and usage of their potential not only for decreasing high level waste volume but also increasing PR by accumulation of even number isotopes ^{236}U and ^{238}Pu. According to Kessler, even for high technology nuclear explosives, ^{238}Pu content between 6-8 wt% would be effective for not to be useful in a nuclear explosive device, owing to the fact that ^{238}Pu develops crucially high alpha heat power compared to other Pu isotopes.[11] In order to validate the feasibility of producing denatured Pu using ERU two different types of codes, MCNP6.2 and MARC, were used. MCNP6.2 was used for neutronics assessment by using the KCODE and BURN feature. Following fuel burnup until 33GWd/MTU and 1 year of cooling, the spent fuel went through reprocessing to separate U isotopes from Pu and fission products then multiple U isotopes were proceeded to a ^{235}U 4.35 wt% by using MARC simulation code to be burned as a fuel. As recycling was repeated, it was observed that ^{236}U component was drastically increased, thus enabling accumulation of ^{238}Pu and it is proper to indicate that using intrinsic features of spent fuel enables denaturing of plutonium.

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