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ENHANCING ADVANCED CANDU PROLIFERATION RESISTANCE FUEL WITH MINOR ACTINIDES

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ABSTRACT

The advanced nuclear system will significantly advance the science and technology of nuclear energy systems and to enhance the spent fuel proliferation resistance. Minor actinides (MA) are viewed more as a resource to be recycled, and transmuted to less hazardous and possibly more useful forms, rather than simply disposed of as a waste stream in an expensive repository facility. MAs can play a much larger part in the design of advanced systems and fuel cycles, not only as additional sources of useful energy, but also as direct contributors to the reactivity control of the systems into which they are incorporated. In this work, an Advanced CANDU Reactor (ACR) fuel unit lattice cell model with 43 UO_2 fuel rods will be used to investigate the effectiveness of a Minor Actinide Reduction Approach (MARA) for enhancing proliferation resistance and improving the fuel cycle performance. The main MARA objective is to increase the ^{238}Pu / Pu isotope ratio by using the transuranic nuclides (^{237}Np and ^{241}Am) in the high burnup fuel and thereby increase the proliferation resistance even for a very low fuel burnup. As a result, MARA is a very effective approach to enhance the proliferation resistance for the on power refueling ACR system nuclear fuel.

The MA transmutation characteristics at different MA loadings were compared and their impact on neutronics criticality assessed. The concept of MARA, significantly increases the ^{238}Pu/Pu ratio for proliferation resistance, as well as serves as a burnable absorber to hold-down the initial excess reactivity. It is believed that MARA can play an important role in atoms for peace and the intermediate term of nuclear energy reconnaissance.

Key Words: Plutonium, Minor Actinide, ACR, CANDU, Proliferation Resistance, Fuel Cycle

1. INTRODUCTION

Key aspects of the advanced nuclear system are: (1) a proliferation-resistant process to separate usable elements in spent nuclear fuel; (2) the reduction of plutonium and minor actinides; and (3) an advanced fuel cycle nuclear system. To accomplish these goals, both international cooperation and public acceptance are crucial. Planned efforts involve near-term and intermediate-term improvements for fuel utilization and recycling in current Light Water Reactors (LWR) as well as the Heavy Water moderated Advanced CANDU Reactors (ACR) that offer much improved fuel utilization and proliferation resistance, along with continued advances in operational safety.

The challenges are solving the energy needs of the world, protection against nuclear proliferation, the problem of nuclear waste, and the global environmental problem. To reduce the amount of spent fuel for storage and enhance proliferation resistance for the intermediate-term, there are two major approaches to consider (a) increase the burnup levels for discharged spent fuel in advanced LWR and ACR (Gen-III

Plus) to reduce spent fuel for storage, (b) use of transuranic nuclides (237Np and 241Am) in high burnup fuel, which can significantly increase the 238Pu/Pu ratio and enhance the proliferation resistance. ACR-1000 is the next-generation (Gen-III Plus) CANDU technology from Atomic Energy of Canada Ltd. (AECL), which maintains proven elements of the existing CANDU design. The ACR-1000 fuel uses slightly enriched uranium (about 2.3%) to extend fuel life to three times so that the spent fuel waste volume is reduced by two-thirds. In this work, we proposed to mix the ACR-1000 (on power refueling operation) fuel with the transuranic nuclides (^{237}Np and ^{241}Am), which can significantly increase the ^{238}Pu/Pu ratio and proliferation resistance.

2. MINOR ACTINIDES REDUCTION APPROCH IN ACR

Issues of nuclear waste and proliferation are directly related to the fuel cycle. The Minor Actinides Reduction Approach (MARA), by mixing the minor actinides with fuel can:, (1) reduce the MA storage volume, (2) enhance the proliferation resistance, and (3) serve as a burnable absorber to improve fuel cycle performance.

The overall goal of proliferation resistance is to prevent the extraction of nuclear materials from civilian nuclear power applications that could be used in the production of nuclear weapons. Based on critical mass considerations, the ^{235}U enrichment limit for proliferation resistance is 20 wt%. However, unlike uranium, any isotopic mix of plutonium has a finite critical mass, i.e., a potential explosive material. Hence, there is no general isotopic concentration threshold for plutonium isotopes from a critical mass point of view. Nevertheless, the suitability for weapons usage varies significantly for plutonium isotopes. Table I, reproduced from Ref. 1, lists the important characteristics of plutonium isotopes. ^{238}Pu, ^{240}Pu, and ^{242}Pu have high spontaneous neutron generation, which reduces the bomb yield significantly. ^{238}Pu also has a high decay heat, which further complicates the design of explosive devices. Consider MARA, where burning minor actinides of ^{237}Np and/or ^{241}Am in the high burnup fuel can transmute to MA which decay to ^{238}Pu in LWRs, which is also the subject of the Protected Plutonium Production (P^3) approach. The subject of the P^3 approach, which was first proposed by Prof. Saito at Tokyo Tech., Japan, can dramatically increase the ^{238}Pu/Pu ratio and enhance the proliferation resistance through the use of a rather heavy load of ^{237}Np (2 wt%).[2] However, ^{237}Np is a controlled nuclear sensitive material. In this study, we use only 0.2 wt% ^{237}Np and/or ^{241}Am to achieve proliferation resistance and improve long fuel cycle performance.

TABLE I. Pu isotope properties important to proliferation resistance.[1]

Isotope	Half-life (years)	Spontaneous Fission Neutrons (n/kg/sec)	Decay Heat (Watt/kg)	Bare Critical Mass (kg)
Pu-238	87.7	2,600,000	560	10
Pu-239	24,100	22	1.9	10
Pu-240	6,560	910,000	6.8	40
Pu-241	14.4	49	4.2	10
Pu-242	376,000	1,700,000	0.1	100

For future advanced nuclear systems, the MAs are viewed more as a resource to be recycled, or transmuted to less hazardous and possibly more useful forms, rather than simply as a waste stream to be disposed of in an expensive repository facility. As a result, they play a much larger part in the design of advanced systems and fuel cycles, not only as additional sources of useful energy, but also as direct contributors to the reactivity control of the systems into which they are incorporated. Fig. 1 shows the MA buildup and decay chains that are most commonly considered in the design of advanced reactors and fuel cycles. As shown in Fig. 1, ^{237}Np and ^{241}Am can be transmuted and decayed to the highly proliferation resistant isotope ^{238}Pu.

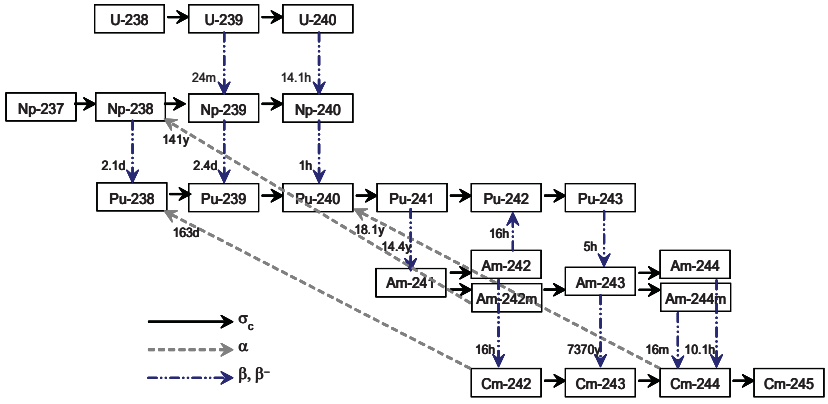

Figure 1. Buildup and decay chains for the MAs. Shaded boxes represent materials with long half-lives that make them of particular interest for transmutation.

In the following study, a typical ACR fuel unit lattice with 43 UO_2 fuel rods model will be used to investigate the effectiveness of MARA for enhancing proliferation resistance and improving the fuel cycle performance in the intermediate term for future nuclear energy systems.

3. ACR-1000 UNIT LATTICE CELL MODEL AND MARA STUDY CASES

A typical ACR fuel unit lattice channel, which contains 12 fuel bundles, with a lattice pitch of 24 cm, as shown in Fig. 2, has been chosen as the basis for the fuel neutronics analysis of UO_2, NpO_2, and AmO_2 at 95% of theoretical density. The fuel rods have a radius of 0.675 (R1 and R2) and 0.575 cm (R3 and R4) and are clad with 0.141 cm of Zr. The 43 fuel rods are arranged in 4 Rings (R1 to R4) as shown in Fig. 2. The center fuel rod (natural U, ^{235}U enrichment 0.71 wt%) contains 4.6 wt% of dysprosium (Dy) absorber. The detailed pressure tube (PT), CO_2 gap, and calandria tube parameters are tabulated in Table II.

TABLE II. ACR-1000 unit lattice cell parameters.

Lattice Cell Parameter	Dimension (cm)	Rod	(cm)
R1 (Center rod)	0.00	OR	0.675
R2 (7 rods)	1.75	OR	0.675
R3 (14 rods)	3.14	OR	0.575
R4 (21 rods)	4.50	OR	0.575
PT IR	5.30	OR	5.95
Gap CO_2 IR	5.95	OR	8.25
Calan. T IR	8.25	OR	8.70
Lattice D_2O pitch	12		
Cladding thickness	0.141		
Effective fuel length	490		
Lattice pitch	24		

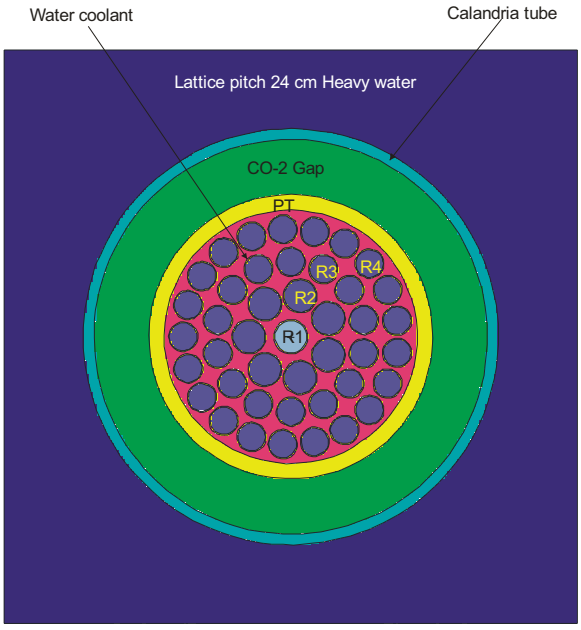

Figure 2. Typical ACR-1000 lattice unit cell model with a lattice pitch of 24 cm.

Increasing the fuel discharge burnup can improve the proliferation resistance and reduce the spent fuel storage volume. In this study, UO_2 with ^{235}U enrichment of 2.3 wt% was used. For the high burnup fuel with the ^{235}U enrichment of 2.3 wt%, three mixed oxide (MO) MA cases for UO_2+NpO_2, UO_2+AmO_2, and UO_2+NpO_2+AmO_2 were established. The ^{235}U enrichment, NpO_2, and AmO_2 composition of the 4 study cases are summarized in Table III.

TABLE III.UO_2 - ^{235}U enrichment, NpO_2, and AmO_2 composition of the 4 study cases.

ID	UO_2 - ^{235}U enrichment (wt%)	NpO_2 (wt %)	AmO_2 (wt%)
Case-1	2.3	--	--
Case-2	2.3	0.2	--
Case-3	2.3	--	0.2
Case-4	2.3	0.12	0.10

4. MONTE CARLO BURNUP METHOD - MCWO

The physics analyses were performed using the computer code MCNP.[3] In addition, the validated fuel burnup methodology MCNP coupled with ORIGEN2,[4] or MCWO,[5] was used. MCWO has been verified at the Idaho National Laboratory (INL) by benchmarking calculated flux magnitudes with measured flux levels for several experiments and in several test positions of the ATR core.[6,7] Computer codes MCNP, MCWO, and ORIGEN2 are contained in the INL listing of qualified codes.

5. RESULTS AND DISCUSSION

MCWO-calculated results for all four case studies will be discussed herein. The nominal power per unit lattice channel is 6.2 MW. The first burnup time interval is 5 effective full power days (EFPDs). The rest of the time intervals are 15 EFPDs up to 740 EFPDs. For each time step, an MCNP KCODE calculation with 20000 source neutrons for 100 cycles is run, requiring ~30 minutes of CPU time on a workstation with two dual-core 2.86 GHz XEON processors. The fission tally calculation for each fuel node can achieve a 1σ standard deviation of 0.5% or less.

The MCWO-calculated K-inf versus burnup for Cases-1 to -4 are plotted in Fig. 3. For K-inf = 1.0, Fig. 3 shows that the discharged burnup of all four cases can reach 22 GWd/t. The higher burnup UO_2 fuel with ^{235}U 2.3 wt% can reduce the spent fuel volume proportionally, which benefits spent fuel storage concerns. From the Fig. 3, it clearly shows that MARA mixed fuel can hold down the initial excess reactivity. The best fuel cycle performance is Case-3 with AmO_2 0.2 wt%, which not only can hold down the initial excess reactivity, but also keeps the K-inf to a very desirable flat profile versus burnup. As a result, the ^{241}Am can also serve as a burnable absorber to effectively hold down the initial excess reactivity (K-inf) from 1.28 to 1.16.

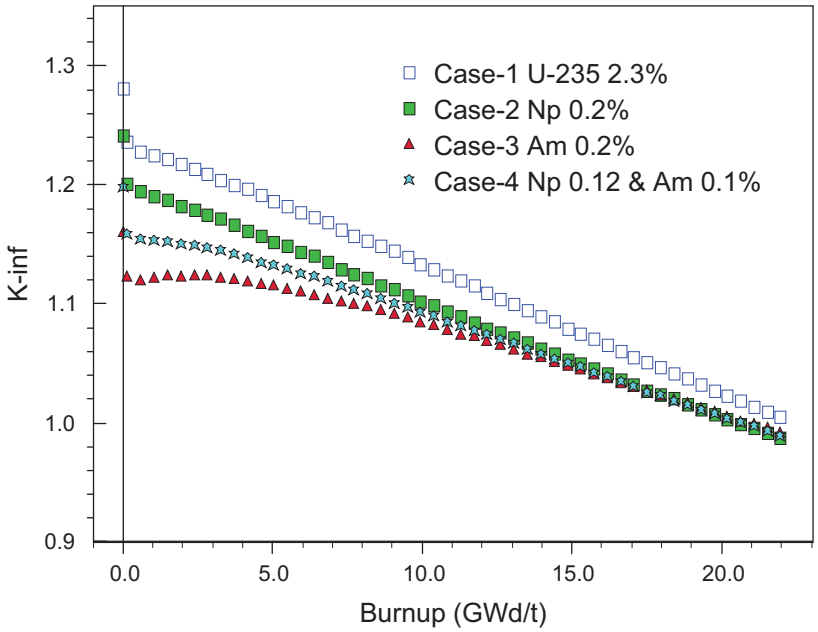

Figure 3. ACR-1000 unit lattice model MCWO-calculated K-inf versus burnup comparison for Cases-1 to -4.

One of the criteria in the definition of spent fuel standards, as defined by the National Academy of Sciences[8] is that the isotopic compositions of the discharged fuel should be about the same as the light water reactor UO_2 spent fuel, particularly, the ^{240}Pu/Pu ratio should be greater than 24%. The MCWO-calculated (43 rods averaged) ^{240}Pu/Pu, ^{238}Pu/Pu, and ^{239}Pu/Pu ratio profiles versus burnup are shown in Figs. 4, 5, and 6, respectively. The MCWO-calculated ^{240}Pu/Pu ratios for Case-1 at the discharged burnup (22 GWd/t) can reach about 28% as shown in Fig. 4. Note, the first MCWO-calculated data point is at 5 EFPDs in Fig. 4, 5, and 7. The MCWO-calculated ^{240}Pu/ Pu ratios for Cases-2, -3, and -4 at the discharged burnup level-off at 25%, 23%, and 24%, respectively. Although, the ^{240}Pu/Pu ratios are less than 27.8% (Case-1) for Cases-2, -3, and -4, at the discharged burnup, the proliferation resistance ^{238}Pu/Pu ratios are considerably higher than the Case-1 ratio at the discharged burnup.

The MCWO-calculated ^{238}Pu/Pu ratio profiles versus burnup are shown in Fig. 5. Fig. 5 shows that the fraction of ^{238}Pu in the fuel increases with burnup, which can better enhance proliferation resistance. Fig. 5 also shows that the fraction of ^{238}Pu in Case-2 dramatically increases to about 9.5%, then, levels off at 8.7%, due to the short β-decay time (2.1 d) for ^{238}Np (see Fig. 1). For Case-3, the transmutation of the ^{241}Am chain, with the long α-decay time (162.8 d) of ^{242}Cm, causes the fraction of ^{238}Pu to reach 10.6% at a burnup of 13.5 GWd/t, then, decreases to about 10%. For Case-4, the transmutation of the ^{237}Np and ^{241}Am chain with the long α-decay time of ^{242}Cm causes the fraction of ^{238}Pu to reach a peak of 10.4% at a burnup of 14 GWd/t, then, decrease to about 9.7%. In summary, Fig. 5 shows that the fraction of ^{238}Pu/Pu of the discharged fuel in Cases-2, -3, and -4 level off about 9.5%, 10%, and 9.7%, respectively, which are all higher than the Case-1 of 0.65%. We conclude that the discharged spent fuel of Case-2, -3 and -4 can effectively enhance proliferation resistance. The combined fractions of ^{238}Pu and ^{240}Pu can meet the spent fuel standard. In addition, G. Kessler[9] pointed out that for ^{238}Pu/Pu above 6%, proliferation resistance can be considered as effective as ^{235}U < 20% or ^{233}U < 12%.

There is a concern that at the low burnup (~35 EFPDs) the ^{240}Pu/ Pu ratio of Case-1 is 3.5%, which is less than the weapons-grade ^{240}Pu/ Pu ratio of 6.5%. However, due to the short decay time (2.1-day) from

^{238}Np to ^{238}Pu, MARA can provide a high fraction of ^{238}Pu at the very low burnup while providing adequate proliferation resistance. For the demonstration, we will subdivide the first time interval of 8 EFPDs into 0.25 EFPD intervals, and the subsequent 35 EFPD time interval into 1 EFPD intervals. MCWO-calculated ^{238}Pu/Pu ratios versus EFPDs is plotted in Fig. 6, which shows that the Case-2 and Case-4 can provide a very high fraction of ^{238}Pu to safeguard the Pu at a very low fuel burnup. To protect Pu in a very low burnup fuel, we loaded Np and Am to a rather heavy 0.2wt%. The penalty for the heavy loaded Np / Am is it can generate more Pu in the discharged fuel. The Pu/U ratio at the discharged burnup (21 GWd/t) for Cases 1 to 4 are 0.61, 0.69, 0.73, and 0.72%, respectively, which indicates the Pu generated are little higher in Cases-2 to 3 than in Case-1.

For reference, the MCWO-calculated ^{239}Pu/Pu ratio profiles versus burnup are shown in Fig. 7, which shows that MARA Cases-2, -3 and -4 have a relatively lower ^{239}Pu/Pu ratio (52 atom%) than the UO_2 Cases-1 and -2 (57 atom%).

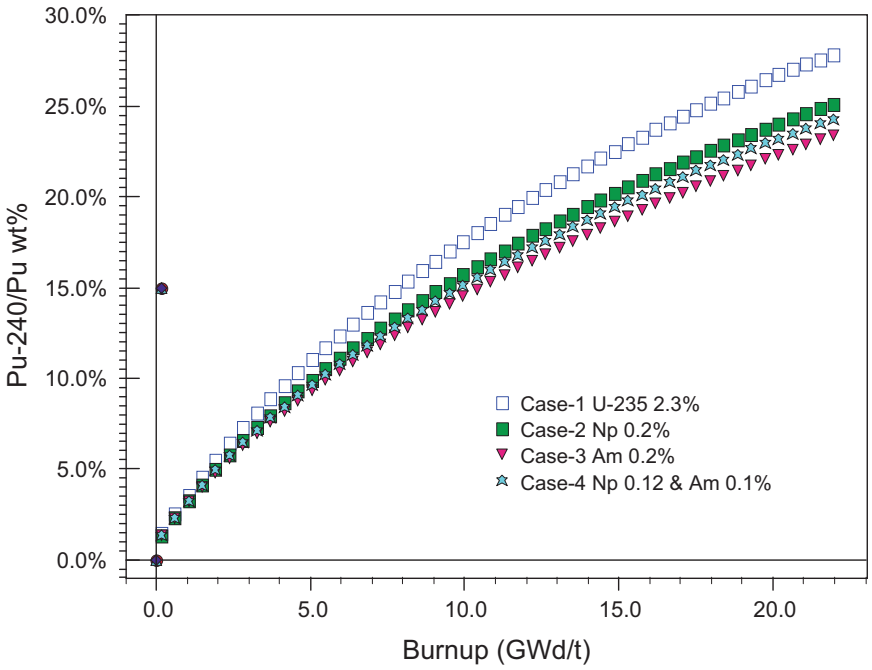

Figure 4. ^{240}Pu/Pu ratio profiles comparison of Cases-1 to -4 versus burnup.

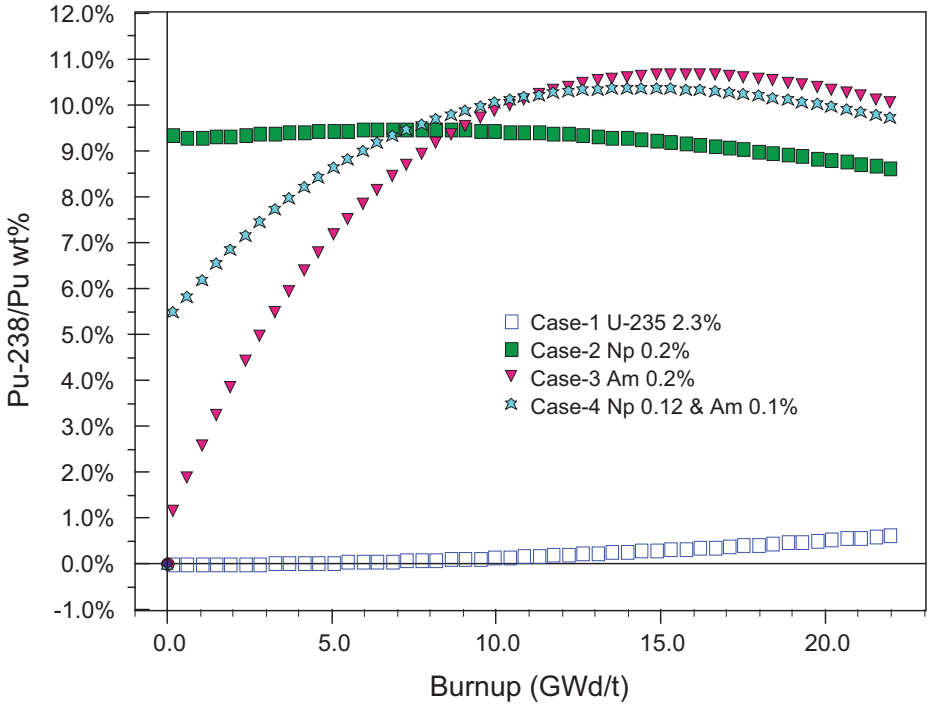

Figure 5. ^{238}Pu/Pu ratio profiles comparison of Cases-1 to -4 versus burnup.

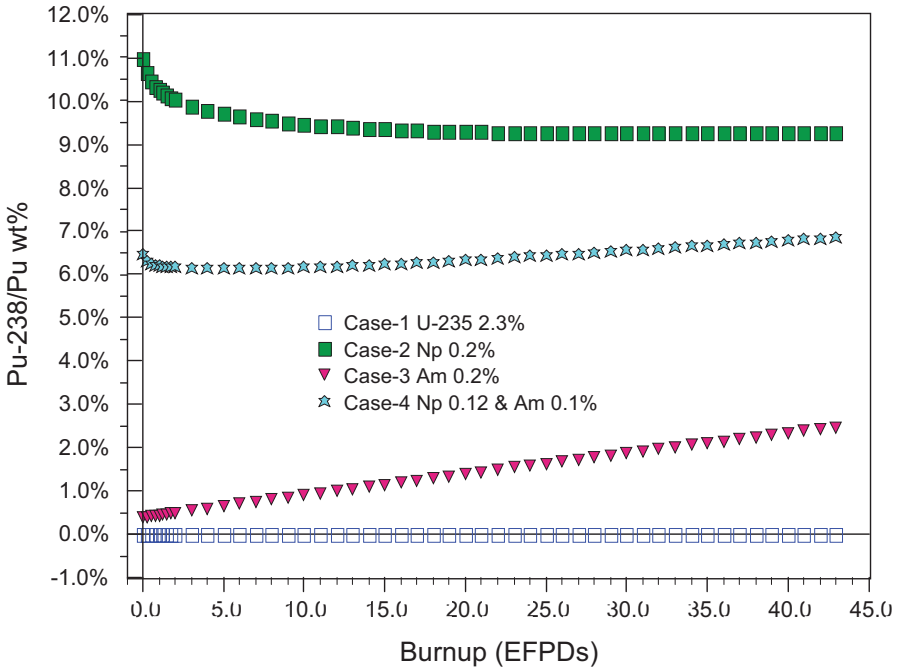

Figure 6. ^{238}Pu/Pu ratio profiles comparison of Cases-1 to -4 versus EFPDs.

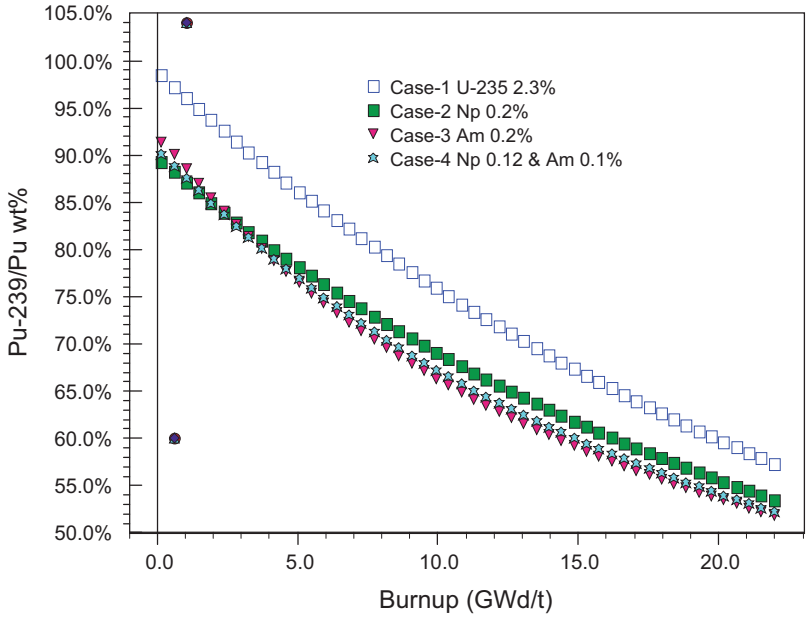

Figure 7. ^{239}Pu/Pu ratio profiles comparison of Cases-1 to -4 versus burnup.

6. CONCLUSIONS

Based on the studies presented herein, it is strongly believed that the concept of MARA, involving the use of transuranic nuclides (^{237}Np and/or ^{241}Am), can significantly increase the ^{238}Pu/Pu ratio for proliferation resistance. ^{241}Am not only can increase the fraction of ^{238}Pu, but also can be used as a burnable absorber to reduce the initial excess reactivity and as serve as a burnable absorber to hold-down the initial excess reactivity. The Case-2 and Case-4 can effectively safe-guard Pu generated in a very low burnup fuel, which is a very important issue in the ACR on power refueling nuclear system. It is believed that MARA can play an important role in atoms for peace and the intermediate term of nuclear energy reconnaissance.

There is a concern that ^{237}Np is a controlled nuclear sensitive material. To address this concerns, it is believed that Case-3 ^{235}U 2.3-wt% with ^{241}Am 0.2-wt% is the best candidate MARA fuel, which not only can achieve the high burnup design goal, but also can achieve the proliferation resistance enhancement goal.

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