Enhancing BWR proliferation resistance fuel with minor actinides

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A B S T R A C T

To reduce spent fuel for storage and enhance the proliferation resistance for the intermediate-term, there are two major approaches (a) increase the discharged spent fuel burnup in the advanced light water reactor- LWR (Gen-III Plus), which not only can reduce the spent fuel for storage, but also increase the ^{238}Pu isotopes ratio to enhance the proliferation resistance, and (b) use of transuranic nuclides (^{237}Np and ^{241}Am) in the high burnup fuel, which can drastically increase the proliferation resistance isotope ratio of ^{238}Pu/Pu. For future advanced nuclear systems, 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. As a result, MAs 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 the study, a typical boiling water reactor (BWR) fuel unit lattice cell model with UO$_2$ fuel pins will be used to investigate the effectiveness of minor actinide reduction approach (MARA) for enhancing proliferation resistance and improving the fuel cycle performance in the intermediate-term goal for future nuclear energy systems. To account for the water coolant density variation from the bottom (0.76 g/cm^3) to the top (0.35 g/cm^3) of the core, the axial coolant channel and fuel pin were divided to 24 nodes. The MA transmutation characteristics at different elevations were compared and their impact on neutronics criticality discussed. The concept of MARA, which involves the use of transuranic nuclides (^{237}Np and/or ^{241}Am), 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.

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1. Introduction

Key aspects of the global nuclear energy partnership (GNEP) are to significantly advance the science and technology of nuclear energy systems and the advanced fuel cycle (AFC) program. GNEP key elements 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. It consists of both innovative nuclear reactors and innovative research in separation and transmutation.

The challenges are solving the energy needs of the world, protection against nuclear proliferation, the problem of nuclear waste, and the global environmental problem. The merits of nuclear energy are the high-density energy, and low environmental impacts i.e. almost zero greenhouse gas emission. The new deployment of nuclear energy reconnaissance is the key to a secure economy and environment. Nuclear energy is clearly needed for future solutions. 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) increasing the burnup levels to reduce spent fuel for storage, (b) using of transuranic nuclides (^{237}Np and ^{241}Am) in high burnup fuel, which can significantly increasing the ^{238}Pu/Pu ratio to enhance the proliferation resistance.

2. Minor actinides reduction approach in LWR

Issues of nuclear waste and proliferation are directly related to the fuel cycle. 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. Ref. [1], lists the important characteristics of plutonium isotopes. ^{238}Pu, ^{240}Pu, and ^{242}Pu have high spontaneous neutron generation, which reduces the

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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 MA which decay to ^{238}Pu in LWRs, which is also the subject of Protected Plutonium Production (P^3) approach. The subject of P^3 approach, which was first proposed by Professor Saito at Tokyo Tech., Japan, can drastically increase the ^{238}Pu/Pu ratio and enhance the proliferation resistance through the use of a rather heavy loaded ^{237}Np (2 wt%) [2]. However, ^{237}Np is a controlled nuclear sensitive material. In this study, we use only 0.5 wt% ^{237}Np and/or ^{241}Am to achieve proliferation resistance and improve long fuel cycle performance.

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. ^{237}Np and ^{241}Am can be transmuted and decayed to the highly proliferation resistant isotope ^{238}Pu.

In the following study, a typical boiling water reactor (BWR) fuel unit lattice cell model with UO$_2$ fuel pins will be used to investigate the effectiveness of MARA for enhancing proliferation resistance and improving the fuel cycle performance in the intermediate-term goal for future nuclear energy systems.

3. BWR unit lattice cell model and MARA study cases

A typical BWR (10 × 10) unit lattice cell, as shown in Fig. 1, has been chosen as the basis for the fuel neutronics analysis of UO$_2$, NpO$_2$, and AmO$_2$ with 95% of theoretical density. The fuel rods have a radius of 0.409 cm and are clad with 0.063 cm of Zr. The fuel pins are arranged in a square fuel lattice. The detailed lattice cell parameters are tabulated in Table 1. The unique feature of the BWR is that the moderator water density decreases from bottom to top
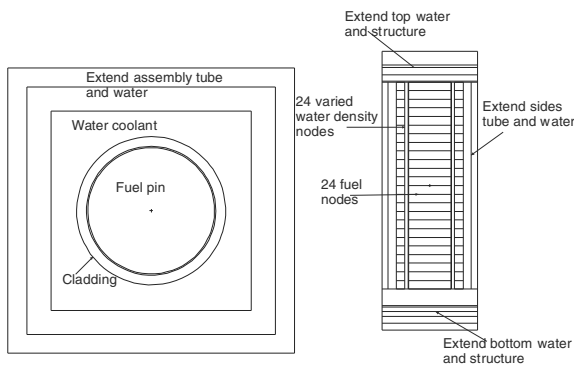

Fig. 1. Typical BWR lattice unit cell model with a fuel pin pitch of 1.25 cm.

Table 1
BWR (10 × 10) unit lattice cell parameters.

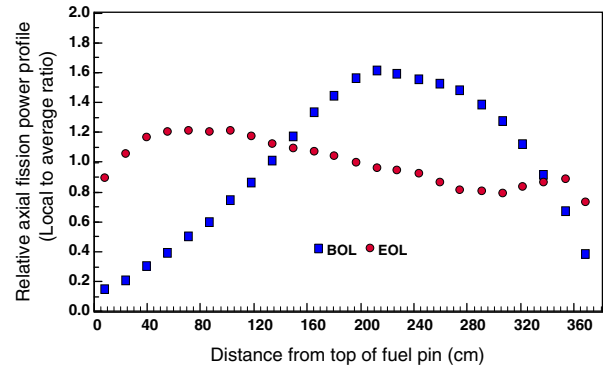| Lattice cell parameter	Dimension (cm)
Extend lattice sides tube and water pitch	1.85
Lattice pitch	1.25
Fuel pin outer radius	0.409
Cladding thickness	0.063
Effective fuel length	376
Extend top water and structure	52
Extend bottom water and structure	72

Fig. 2. BWR (10 × 10) unit lattice, the relative 24 axial nodes fission power profiles at BOL and EOL.

Table 2
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	4.95	–	–
Case-2	4.95	0.5	–
Case-3	4.95	–	0.5
Case-4	4.95	0.25	0.25

of core. We divided the water coolant channel and fuel pin into 24 axial nodes as shown in Fig. 2. The relative fission power local to average ratio (L2AR) profiles along the 24 fuel nodes at the beginning of life (BOL) and at the end of life (EOL) at the discharged burnup of 50 GWd/t (see Section 5) are shown in Fig. 2.

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 4.95 wt% was used. For the high burnup fuel with ^{235}U enrichments of 4.95 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 2.

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. 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 burnup time interval is 1.25 GWd/t. For each time step, an MCNP KCODE calculation with 8000 source neutrons for 100 cycles is run, requiring ~15 min 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 2% 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. 4 shows that the discharged burnup of all four cases can reach 50 GWd/t. The higher burnup UO$_2$ fuel with ^{235}U 4.95 wt% can reduce the spent fuel volume proportionally,
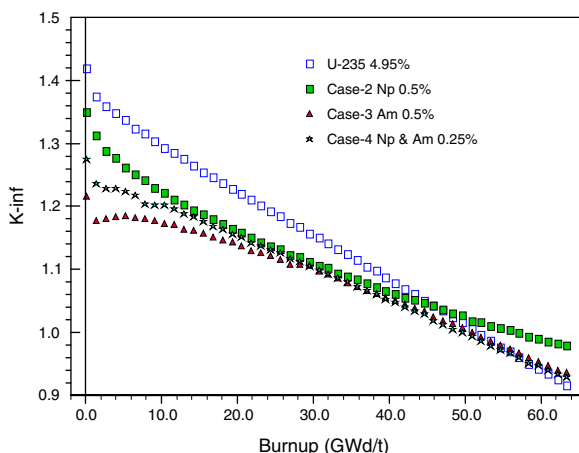
Fig. 3. BWR lattice unit cell MCWO-calculated K-inf versus burnup comparison for Cases-1 to -4.
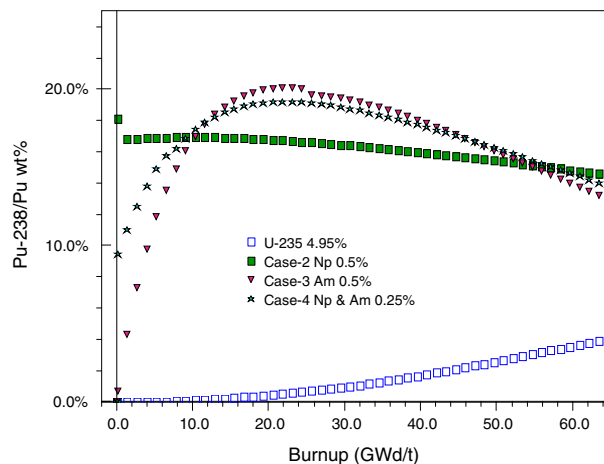

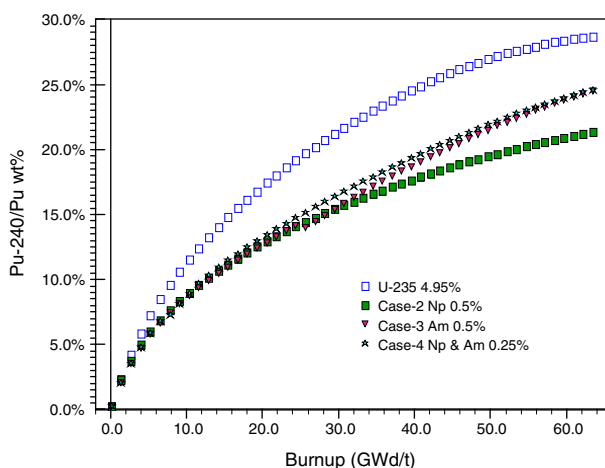Fig. 5. ^{238}Pu/Pu ratio profiles comparison of Cases-1, -2, and -3 versus burnup.

Fig. 4. ^{240}Pu/Pu ratio profiles comparison of Cases-1, -2, and -3 versus burnup.

which benefits the 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$_2$ 0.5 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 serve as a burnable absorber to effectively hold down the initial excess reactivity (K-inf) from 1.42 to 1.22.

One of the criteria in the definition of spent fuel standard, as defined by the National Academy of Sciences [6] 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 (24 nodes averaged) ^{240}Pu/Pu and ^{238}Pu/Pu ratio profiles versus burnup are shown in Figs. 4 and 5, respectively. The MCWO-calculated ^{240}Pu/Pu ratios for Case-1 at the discharged burnup (50 GWd/t) can reach about 27% as shown in Fig. 4. The MCWO-calculated ^{240}Pu/Pu ratios for Cases-2, -3, and -4 at the discharged burnup are level-off at 20%, 22%, and 23%, respectively. Although, the ^{240}Pu/Pu ratios are marginally less than 24% for Cases-2, -3, and -4, at the discharged burnup, the proliferation resistance ^{238}Pu/Pu ratios are considerably higher than the Case-1 3% at the discharged burnup. As a result, the (^{238}Pu + ^{240}Pu)/Pu ratios are larger than 24%.

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 drastically increases to about 17%, then, levels off at 15%, due to the short β-decay time (2.1 day) for ^{238}Np. For Case-3, the transmutation of the ^{241}Am chain, with the long α-decay time of ^{242}Cm, causes the fraction of ^{238}Pu to peak at 20% at a burnup of 22 GWd/t, then, decreases to about 15%. 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 19% at a burnup of 22 GWd/t, then, decrease to about 16%. In summary, Fig. 5 shows that the fraction of ^{238}Pu of the discharged fuel in Cases-2, -3, and -4 level off about 15%, 15%, and 16%, respectively, which are all higher than the Case-1 of 3%. We conclude that the discharged spent fuel of Cases-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, Kessler [7] 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 (∼10 GWd/t) the ^{240}Pu/Pu ratios are less than 6.5%, which qualify as weapons-grade Pu. To protect Pu in the low burnup fuel, we loaded Np and Am rather heavy as of 0.5 wt%, such that MARA can provide a high fraction of ^{238}Pu at the low burnup while providing adequate proliferation resistance. The disadvantage of the heavy loaded Np/Am can generated more Pu in the discharged fuel. The Pu/U ratio at the discharged burnup (50 GWd/t) for Cases-1 to -4 are 0.97%, 1.54%, 1.34%, and 1.30%, respectively, which indicates the Pu generated are higher in Cases-2 to -3 than in Case-1. Additional cost of MARA R&D is another disadvantage.

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, as well as serve 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.

The known disadvantages of MARA are: (a) ^{237}Np and ^{241}Am in the fuel will generate more [4]. He, which will affect the fission gas release performance; (b) The need to have improved transuranic

cross-section library data; (c) The ^{237}Np is a controlled nuclear sensitive material.

And, the encouraging advantages of MARA are: (a) The high burnup of the spent fuel (^{235}U 4.95 wt%) is well developed and becoming a standard for LWR fuel reloading, which can effectively reduce the spent fuel storage volume; (b) ^{237}Np and/or ^{241}Am can drastically increase the fraction of ^{238}Pu to enhance the proliferation resistance; (c) ^{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; (d) ^{237}Np can be transmuted quickly with β-decay (half-life 2.1 days) to a high fraction of ^{238}Pu, which can effectively enhance the proliferation resistance for early removal of low burnup fuel.

As a result of the above concerns, it is believed that Case-3 ^{235}U 4.95 wt% with ^{241}Am 0.5 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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