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Minor Actinides Loading Optimization for Proliferation Resistant Fuel Design - BWR

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Abstract – *One approach to address the United States Nuclear Power (NP) 2010 program for the advanced light water reactor (LWR) (Gen-III+) intermediate-term spent fuel disposal need is to reduce spent fuel storage volume while enhancing proliferation resistance. One proposed solution includes increasing burnup of the discharged spent fuel and mixing minor actinide (MA) transuranic nuclides (^{237}Np and ^{241}Am) in the high burnup fuel. Thus, we can reduce the spent fuel volume while increasing the proliferation resistance by increasing the isotopic ratio of ^{238}Pu/Pu. For future advanced nuclear systems, MAs 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 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.*

A typical boiling water reactor (BWR) fuel unit lattice cell model with UO_2 fuel pins will be used to investigate the effectiveness of adding MAs (^{237}Np and/or ^{241}Am) to enhance proliferation resistance and improve fuel cycle performance for the intermediate-term goal of future nuclear energy systems. However, adding MAs will increase plutonium production in the discharged spent fuel. In this work, the Monte-Carlo coupling with ORIGEN-2.2 (MCWO) method was used to optimize the MA loading in the UO_2 fuel such that the discharged spent fuel demonstrates enhanced proliferation resistance, while minimizing plutonium production. The axial averaged MA transmutation characteristics at different burnup were compared and their impact on neutronics criticality and the ratio of ^{238}Pu/Pu discussed.

I. INTRODUCTION

Key aspects of the United States Nuclear Power (NP) 2010 program for the advanced light water reactor (LWR) (Gen-III+) and Advanced Fuel Cycle programs are: (1) proliferation-resistant processes to separate usable elements from spent nuclear fuel and (2) reduction of spent nuclear fuel storage volume by using high burnup fuel. Advancement of the science and technology of nuclear energy systems consists of both innovative advanced nuclear fuel and innovative research in separation and transmutation. The challenges are solving the energy needs of the world, protection against nuclear proliferation, and the problem of nuclear waste. The concept of mixing MAs with high burnup oxide fuel can dramatically increase the ^{238}Pu/Pu ratio for proliferation resistance,[1] as well as reduce the MAs generated in the LWR discharged spent fuel. It is believed that mixing MAs can play an important role in the atoms for peace application of the advanced nuclear energy system.

To reduce spent nuclear fuel volume and enhance proliferation resistance of LWR Gen-III+ for the intermediate-term, there are two major approaches - (1) increase the burnup levels to reduce spent fuel for storage, (2) mix minor actinide (MA) transuranic nuclides (^{237}Np and ^{241}Am) in the advanced high burnup fuel, thus significantly increasing the ^{238}Pu/Pu ratio and enhancing the proliferation resistance. However, as pointed out in Ref. 1, adding the MAs will increase undesired plutonium production in the discharged spent fuel as much as 1.58 times. In this work, the Monte-Carlo coupling with ORIGEN-2.2 (MCWO) method was used to optimize the MA loading in the UO_2 fuel such that the discharged spent fuel demonstrates enhanced proliferation resistance, while minimizing plutonium production and maintaining a lower, yet meaningful MA reduction rate. The reactor axial averaged MA transmutation characteristics at different burnup levels were compared and their impact on neutronics criticality and the ratio of ^{238}Pu/Pu discussed.

II. OPTIMIZED MINOR ACTINIDE LOADING IN HIGH BURNUP FUEL

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 the different plutonium isotopic compositions. Ref. 2 lists the important proliferation resistance characteristics of plutonium isotopes and summarized in Table 1 of Ref. 1. ^{238}Pu, ^{240}Pu, and ^{242}Pu have high spontaneous neutron generation, which reduces the bomb yield significantly. In addition, ^{238}Pu has high decay heat, which further complicates the design of explosive devices. Because ^{238}Pu is highly proliferation resistant, G. Kessler[3] pointed out that for $^{238}Pu/Pu$ ratios above 6%, its proliferation resistance can be considered as effective as $^{235}U < 20\%$ or $^{233}U < 12\%$.

Burning MA of ^{237}Np and/or ^{241}Am mixed in the LWR high burnup fuel enhances the transmutation of which decay to ^{238}Pu. To protect plutonium even in the low burnup fuel, per Ref. 1, the author loaded Np rather heavily (0.5 wt%), such that the mixing MA provided a high fraction of ^{238}Pu at very low burnup while providing adequate proliferation resistance. The disadvantage of the heavy loading of Np/Am is that more plutonium is generated in the discharged fuel. The $Pu/^{238}U$ ratio at the discharged burnup (51 GWd/t) for UO_2 only and UO_2 mixed with ^{237}Np 0.5 wt% are 0.97 and 1.54,[1] respectively, which indicates the undesired plutonium generation is much higher in the mixed ^{237}Np and UO_2 fuel. In this study, we mix only 0.125 wt% ^{237}Np and/or ^{241}Am in the MA Loading Optimization (MALO) approach to achieve the goal of proliferation resistance as well as minimize the plutonium production in the spent fuel.

For future advanced nuclear systems, the MAs are considered 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, MAs will play a much larger role in the design of advanced systems and fuel cycles, not only as additional sources of useful energy, but also as direct contributors to the safeguard of the generated Pu in the spent fuel. ^{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 MALO approach for enhancing proliferation resistance for future advanced nuclear energy systems.

III. BWR UNIT LATTICE CELL MODEL AND MALO APPROACH

A typical BWR (10x10) unit lattice cell, as shown in Fig. 1 of Ref. 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 II of Ref. 1. The unique feature of a BWR is that the moderator water density decreases from bottom to top of the core. We divided the water coolant channel and fuel pin into 24 axial nodes as shown in Fig. 2 of Ref. 1. The validation of 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) for a discharged burnup of 50 GWd/t are discussed in Ref. 1.

Increasing the fuel discharge burnup can improve the proliferation resistance and reduce the spent fuel storage volume. In this work, 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 (MOX) 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 proposed study cases are summarized in Table I. Case-4 represents the MA optimized loaded fuel, which minimizes plutonium production in the discharged high burnup fuel, while meeting the proliferation resistance goal of $^{238}Pu/Pu$ ratio greater than 6% and maintaining a lower, yet still quite effective MA reduction rate.

TABLE I
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	0.125	--
Case-3	4.95	--	0.125
Case-4	4.95	.025	0.105

IV. MONTE CARLO BURNUP METHOD – MCWO

The physics analyses were performed using the computer code MCWO[4] which couples MCNP[5] calculations with the ORIGEN2.2[6,7] fuel burnup calculations. 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 Advanced Testing Reactor (ATR) core.

At each calculation (burnup) interval, MCNP results are used to compute the fission power distribution and burnup-dependent cross sections for each fuel pin, the updated data are then transferred to ORIGEN2.2 for cell-wise depletion calculations. The MCNP-generated reaction rates are integrated over the continuous-energy nuclear data and space within the region. Any odd or regular shaped region in the MCNP model can be depleted (on average) with reaction rate data that can be more accurate than the few-group data used in the commercial LWR industry. MCNP, MCWO, and ORIGEN2.2 are contained in the INL listing of qualified analysis computer codes.

V. RESULTS AND DISCUSSION

MCWO-calculated results for all four case studies will be discussed. 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 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 2% or less. The MCWO-calculated K-inf versus burnup, the ratio of plutonium isotopes (^{240}Pu/PU and ^{238}Pu/Pu), and the ratio of generated and depleted actinides ^{239}Pu/^{238}U, ^{237}Np/^{238}U, and ^{241}Am/^{238}U, versus burnup are compared and discussed below.

V.A. K-inf versus Burnup

The MCWO-calculated K-inf versus burnup for Cases-1 to -4 are plotted in Fig. 1. For K-inf = 1.0, Fig. 1 shows that the discharged burnup of Cases-1 to -4 can reach 51, 58, 51, and 51 GWd/t, respectively.

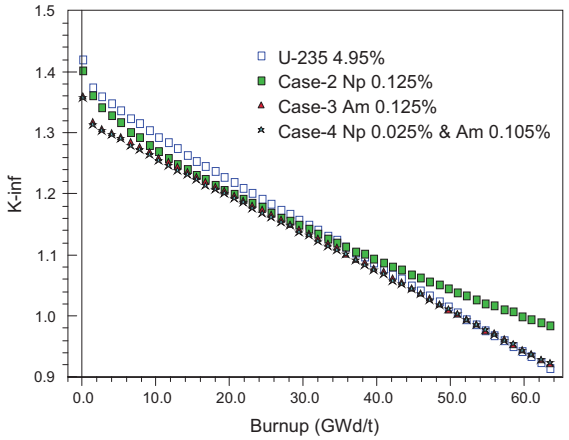

Fig. 1 BWR lattice unit cell MCWO-calculated K-inf versus burnup comparison for Cases-1 to -4.

The higher burnup UO_2 fuel with ^{235}U 4.95 wt% reduces the spent fuel volume proportionally, positively impacting spent fuel storage concerns. Due to the transmutation of ^{237}Np and subsequent decay to ^{238}Pu, Fig. 1 also shows that ^{237}Np mixed UO_2 fuel extends discharged burnup to 58 GWd/t.

V.B. Ratio of Plutonium isotopes versus Burnup

One of the criteria in the definition of spent fuel standard, as defined by the National Academy of Sciences[8] is that the isotopic compositions of the discharged fuel should be about the same as LWR 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. 2 and 3, respectively. The MCWO-calculated ^{240}Pu/Pu ratios for Case-1 at discharged burnup (51 GWd/t) reach about 27% as shown in Fig. 2. The MCWO-calculated ^{240}Pu/ Pu ratios for Cases-2, -3, and -4 at discharged burnup level-off at 24%, 26%, and 25%, respectively. The ^{240}Pu/Pu ratios are marginally larger than 24% for Cases-2, -3, and -4, at discharged burnup, meeting the standard spent fuel criteria.

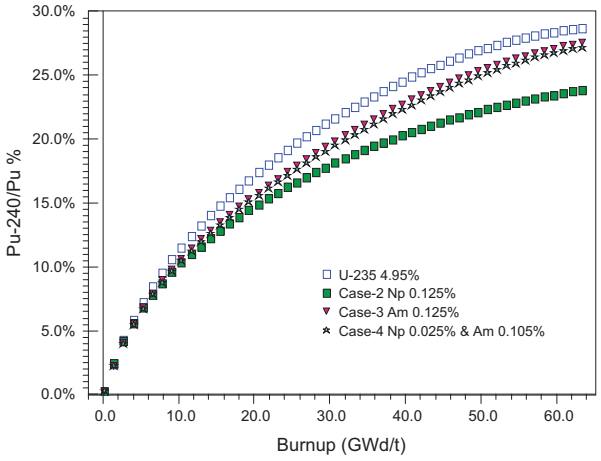

Fig. 2 ^{240}Pu/Pu ratio profiles comparison of Cases-1 to -4 versus burnup.

The MCWO-calculated ^{238}Pu/Pu ratio profiles versus burnup are shown in Fig. 3. Fig. 3 shows that the ratio of ^{238}Pu/Pu in the fuel increases with burnup, enhancing proliferation resistance. The ratios of ^{238}Pu/Pu for Cases-1 to -4 are 2.7%, 6.9%, 6.7%, and 8.0%, respectively, at discharged burnup. Due to the rather short decay half-life (2.1-day) from ^{238}Np to ^{238}Pu, Fig. 3 also shows that the ratio of ^{238}Pu/Pu in Case-2 quickly increases to about 6.3%. For Case-3, the transmutation of the ^{241}Am chain, with the longer α-decay time (163 d) of ^{242}Cm, causes the ratio of ^{238}Pu to peak at 7.3%, then, decrease to about 6.7%. For Case-4, the transmutation of ^{237}Np and ^{241}Am chain with the long α-decay time of ^{242}Cm causes the ratio

of ^{238}Pu to reach a peak of 8.8% at a burnup of 24 GWd/t, then, decrease to about 8.0%. Because of the shorter decay half-life (2.1-day) from ^{238}Np to ^{238}Pu, the added ^{237}Np in Case-4 can reach ^{238}Pu/Pu ratio of 6% at 6.6 GWd/t. Case-3 without ^{237}Np reaches ^{238}Pu/Pu ratio of 6% at 11.7 GWd/t. In summary, Fig. 3 shows that the ratio of ^{238}Pu for the discharged fuel in Cases-2, -3, and -4 are all higher than Case-1 with a ^{238}Pu/Pu ratio of 2.7%. We conclude that the discharged spent fuel of Cases-2, -3, and -4 can effectively enhance proliferation resistance. The proliferation resistance ^{238}Pu/Pu ratios are considerably higher at the discharged burnup for Cases-2, -3, and -4 than the ^{238}Pu/Pu ratio for Case-1. The (^{238}Pu+^{240}Pu)/Pu ratios are much larger than the spent fuel standard of 24%. In addition, G. Kessler[3] pointed out that for ^{238}Pu/Pu ratio above 6%, proliferation resistance can be considered as effective as ^{235}U < 20% or ^{233}U < 12%.

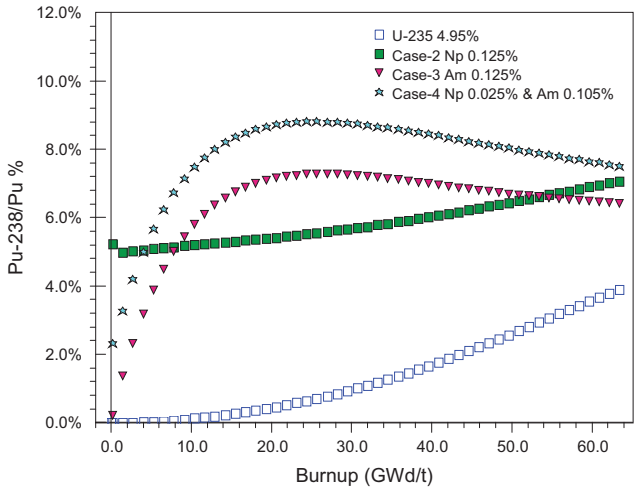

Fig. 3 ^{238}Pu/Pu ratio profiles comparison of Cases-1 to -4 versus burnup.

V.C. Ratio of Generated and Depleted Actinides to Initial ^{238}U versus Burnup

As discussed in Ref. 1, the heavily mixed NpO_2 and AmO_2 in the BWR UO_2 fuel will significantly increase the generated plutonium in the discharged spent fuel. The MCWO-calculated profiles of the ratio of generated plutonium to initial ^{238}U versus burnup for the four cases are plotted in Fig. 4. At the discharged burnup, the Pu/^{238}U ratio versus burnup for Cases-1 to -4 are 0.97%, 1.34%., 1.04%, and 1.06%, respectively. Although, ^{237}Np increases the ^{238}Pu/Pu ratio quickly with Case-2, it will generate 1.38 times more plutonium than reference Case-1. For Case-3 and -4 only 1.08 times more plutonium is generated than Case-1.

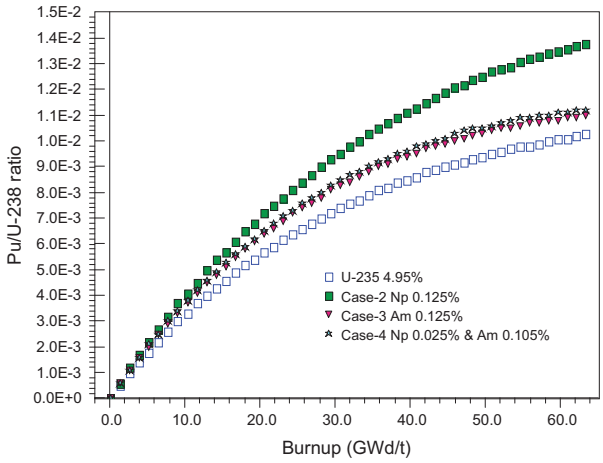

Fig. 4 Generated Pu/^{238}U ratio profiles comparison of Cases-1, -2, -3, and -4 versus burnup.

The MCWO-calculated ratio of the generated and then depleted ^{237}Np to the initial ^{238}U versus burnup with zero cooling time (CT) are plotted in Fig. 5. The generated ^{237}Np/^{238}U ratios for Case-1 and -3, reach about 5.8×10^{-4}. For Case-4, the ^{237}Np/^{238}U ratio at initial loading and then discharged burnup are 5.3×10^{-4} and 7.6×10^{-4}, respectively, which represents a gain of 2.3×10^{-4}. Because of the high ^{237}Np in Case-2, the ^{237}Np/^{238}U ratios at the initial and discharged burnup are 1.3×10^{-3} and 1.1×10^{-3}, respectively.

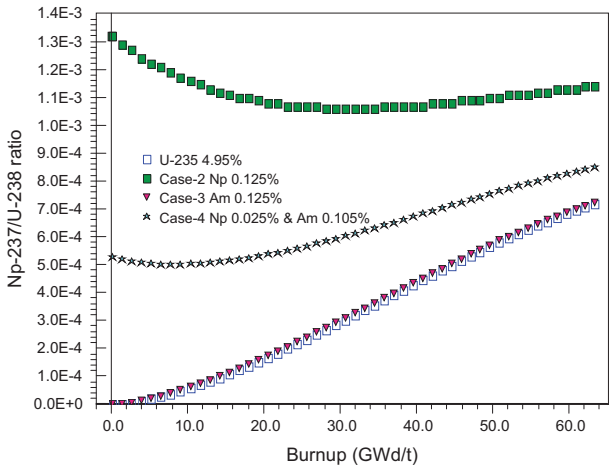

Fig. 5 Generated and depleted ^{237}Np/^{238}U ratio profiles comparison of Cases-1, -2, -3, and -4 versus burnup.

The MCWO-calculated ratio of the generated and then depleted ^{241}Am to the initial ^{238}U versus burnup with zero CT are plotted in Fig. 6. The generated ^{241}Am/^{238}U ratios for Case-1 and -2, reach about 5.3×10^{-5} and 8.3×10^{-5}, respectively. For Case-3, the ^{241}Am/^{238}U ratios at the initial loading and then the discharged burnup are 1.28×10^{-3} and 1.1×10^{-4}, respectively. Similarly, for Case-4, the

$^{241}Am/^{238}U$ ratios at the initial loading and then the discharged burnup are $1.23x10^{-3}$ and $1.1x10^{-4}$, respectively. Because of a rather low loading of ^{241}Am in Case-3, its initial $^{241}Am/^{238}U$ ratio ($1.28x10^{-3}$) approaches the same value ($1.11x10^{-4}$) as Case-1 at the discharged burnup.

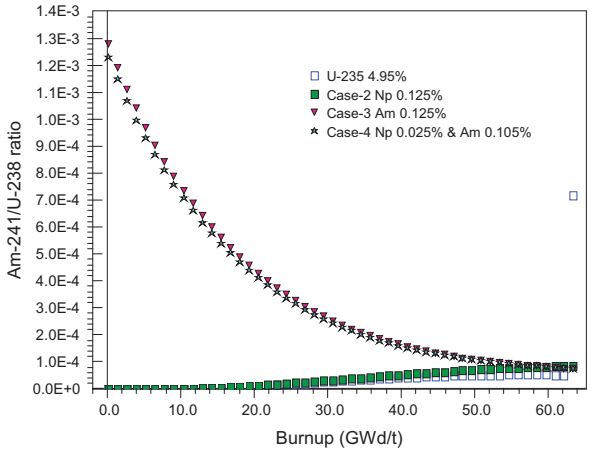

Fig. 6 Generated and depleted $^{241}Am/^{238}U$ ratio profiles comparison of Cases-1, -2, -3, and -4 versus burnup.

V.D. The Impact of Actinide Buildup and Decay with Respect to Cooling Times

All the above results discussed assumed zero cooling time (CT) at the end of each time step. However, the detailed isotope buildup and decay chains for the actinides are quite complex as shown in Fig. 7. The decay half-life of the $^{238,239,240,241,242}Pu$ are, 87.7-years, 24,100-years, 6,560-years, 14.4-years, and 376,000-years, respectively, which will impact the $^{238}Pu/Pu$ and $^{241}Am/^{238}U$ ratio with respect to different cooling time. Note the short half-life of ^{241}Pu, which decays more quickly to ^{241}Am.

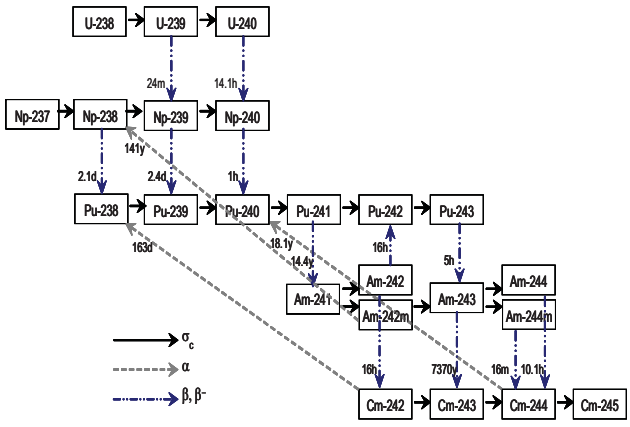

Fig. 7 Buildup and decay chains for the actinides.

In this subsection, we will investigate the impact of different cooling time for the optimized Case-3: $^{238}Pu/Pu$ ratio profiles at the end of each burnup time step. The ORIGEN2.2 calculated $^{238}Pu/Pu$ ratio profiles with zero, one-month, half-year, one-year, 5-year, and 10-year cooling time are plotted at Fig. 8. Because of the long 163-day α-decay time of ^{242}Cm to ^{238}Pu, Fig. 8 indicates that if the cooling time is longer than a half-year, the $^{238}Pu/Pu$ ratio will be greater than the proliferation resistance index of 6%. Therefore, Case-3 can effectively safe-guard the generated plutonium for all burnup with cooling time longer than a half-year.

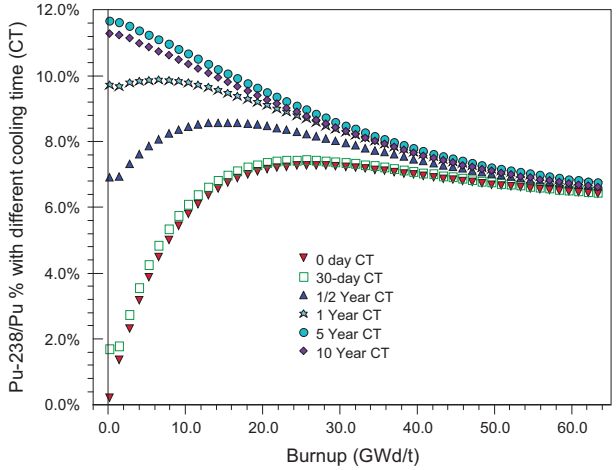

Fig. 8 Case-3 $^{238}Pu/Pu$ ratio profiles with respect to cooling time.

At the discharged burnup (51 GWd/t), the ORIGEN2.2 calculated $^{241}Am/^{238}U$ ratio is about $1.11x10^{-4}$ for both Case-1 and -3. Then, ORIGEN2.2 was used to calculate the discharged $^{241}Am/^{238}U$ ratio versus cooling time for 0-day, 1-day, 30-day, ½-year, 1-year, 2-year, 3-year, 5-year, 10-year, 15-year, and 20-year. The $^{241}Am/^{238}U$ ratio versus cooling time is plotted in Fig. 9. Due to a short half-life of 14.4-years for ^{241}Pu, ^{241}Pu decays to ^{241}Am and for Case-1 and -3 increases the $^{241}Am/^{238}U$ ratio from $1.11x10^{-4}$ to $9.28x10^{-4}$ as shown in Fig. 9. It represents a reduction of $^{241}Am/^{238}U$ ratio from the initial ^{241}Am loading to the generated ^{241}Am at the discharged spent fuel with 20-years of cooling time as $3.52x10^{-4}$ ($1.28x10^{-3}$– $9.28x10^{-4}$) for Case-3. As a result, Case-3 can reduce about 38% ($3.52x10^{-4}$ / $9.28x10^{-4}$) of the BWR generated ^{241}Am in the discharged spent fuel with 20-years of cooling time.

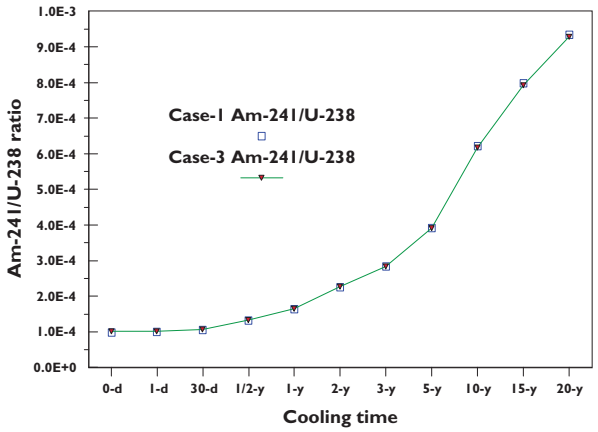

Fig. 9 Cases-1 and -3 decayed and buildup of $^{241}Am/^{238}U$ ratio with respect to cooling time.

VI. CONCLUSIONS AND RECOMMENDATION

Based on these studies, it is strongly believed that the concept of the MALO approach, involving the use of transuranic nuclides (^{237}Np and/or ^{241}Am), significantly increases the $^{238}Pu/Pu$ ratio for proliferation resistance, which effectively safe-guards the generated plutonium to support atoms for peaceful use of nuclear energy.

The ^{237}Np is a controlled nuclear sensitive material. For Case-2, the ^{237}Np generated about 1.35 times more plutonium in the discharged spent fuel than that of Case-1. The added ^{237}Np to Case-4 slightly lowers the burnup (6.6 GWd/t) compared with Case-3 to reach the proliferation resistance index for a $^{238}Pu/Pu$ ratio of 6%. We would conclude that Case-3 ^{235}U 4.95-wt% with ^{241}Am 0.125-wt% is the best candidate MALO fuel, which achieves the high burnup design goal, as well as achieves the proliferation resistance enhancement goal. For the secondary MA reduction goal, Case-3 can reduce a meaningful 38% of a BWR generated ^{241}Am in the discharged spent fuel with 20-years of cooling time.

The high burnup fuel (^{235}U 4.95-wt%) is well developed and becoming a standard for BWR fuel reloading, which can effectively reduce the spent nuclear fuel storage volume. However, the mixed ^{237}Np and ^{241}Am in the MALO fuel will generate more ^{4}He, which will affect fuel fission gas release performance. And, the need to have improved transuranic cross-section library data. We strongly recommend having the designed BWR MALO fuel V&V irradiation testing conducted in the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL).

REFERENCES

1. G. S. CHANG, "Enhancing BWR Proliferation Resistance Fuel with Minor Actinides," Journal of Nuclear Materials, NUMA 43671 to be published (2009).
2. J. C. MARK, "Explosive Properties of Reactor-Grade Plutonium," *Science & Global Security*, **4**, 111-128 (1993).
3. G. KESSLER, "Plutonium Denaturing by ^{238}Pu," *Nucl. Sci. & Eng.*: **155**, 53-73 (2007).
4. G. S. CHANG and J. M. Ryskamp, "Depletion Analysis of Mixed Oxide Fuel Pins in Light Water Reactors and the Advanced Test Reactor," *Nucl. Technol*., Vol. 129, No. 3, 326-337 (2000).
5. T. GOORLEY, J. Bull, F. Brown, et. al., "Release of MCNP5_RSICC_1.30," MCNP Monte Carlo Team X-5, LA-UR-04-4519, Los Alamos National Laboratory, November 2004.
6. A. G. CROFF, "ORIGEN2: A Versatile Computer Code for Calculating the Nuclide Compositions and Characteristics of Nuclear Materials," *Nuclear Technology*, Vol. 62, 335-352 (1983).
7. S. B. Ludwig and A. G. Croff to Jennie Manneschmidt letter report, "Revision to ORIGEN2 – Version 2.2," Oak Ridge National Laboratory, May 23, 2002.
8. National Academy of Sciences Committee on International Security and Arms, Management and Disposition of Excess Weapons Plutonium, National Academy Press, 1994.