Americium Transmutation Feasibility When Used as Burnable Absorbers - 12392

Sean A. Barbaras*, Travis W. Knight**
*United States Military Academy, West Point, New York 10996
** University of South Carolina, Columbia, South Carolina 29208

ABSTRACT

The use of plutonium in Mixed Oxide (MOX) fuel in traditional Pressurized Water Reactor (PWR) assemblies leads to greater americium production which is not addressed in MOX recycling. The transuranic nuclides (TRU) contribute the most to the radiotoxicity of nuclear waste and a reduction of the TRU stockpile would greatly reduce the overall radiotoxicity of what must be managed. Am-241 is a TRU of particular concern because it is the dominant contributor of total radiotoxicity for the first 1000 years in a repository. This research explored the feasibility of transmuting Am-241 by using varying amounts in MOX rods being used in place of burnable absorbers and evaluated with respect to the impact on incineration and transmutation of transuranics in MOX fuel as well as the impact on safety. This research concludes that the addition of americium to a non-uniform fuel assembly is a viable method of transmuting Am-241, holding down excess reactivity in the core while serving as a burnable poison, as well as reducing the radiotoxicity of high level waste that must be managed.

INTRODUCTION

Using MOX fuel in traditional PWR assemblies has been researched at length and has shown to provide the benefit of transmutation and targets the amount and toxicity of high level waste needed to be managed. In this research, fuel assemblies consisting of both MOX and Uranium Dioxide (UO_2) were used in a core model and evaluated using the computer code NESTLE to solve the few-group neutron diffusion equation utilizing the nodal expansion method. Varying weight percents of Am-241 were added to the MOX fuel as burnable absorbers to hold down excess reactivity and shape the power distribution in the core as well as determine the feasibility of transmuting Am-241.

Americium Transmutation

The transuranic nuclides contribute the most to the radiotoxicity of nuclear waste and a reduction of the TRU stockpile would greatly reduce the overall radiotoxicity of what must be managed. Americium is a TRU of particular concern because it is the dominant contributor of total radiotoxicity for the first 1000 years in a repository. Spent UO_2 fuel that has been burned to 55000MWD/MTU originally enriched to 4.9w/o U-235 contains 0.012017w/o americium with three isoptopes (0.003822w/o Am-241, 0.000074w/o Am-242m, and 0.008121w/o Am-243) [1]. Considering the actinides from PWR type reactors from many years of accumulation, Am-241 makes up 90% of the total radiotoxicity of the required repository storage [2]. Therefore, the main purpose behind the transmutation of americium is to transmute Am-241, which is the isotope focused on in this research.

The transmutation of Am-241 takes advantage of the thermal energy large capture cross-sections of 800 barns as well as the very large fission cross-section of 3000 barns of the short lived daughter isotope, Am-242 [4]. Americium is not found in nature but is produced by neutron capture reactions by plutonium isotopes. Am-241 is produced as Pu-241 decays by beta particle emission and Pu-241 is produced from the successive neutron absorption of Pu-239. Am-241

has a half-life of 430 years and decays by emitting an alpha particle [3]. In order to reduce the amount of Am-241 that must be managed as nuclear waste, this isotope can be transmuted.

METHOD

CORAIL Assembly

Because of the radiotoxicity problems associated with americium, this research included the feasibility of transmuting Am-241 using non-uniform hybrid Am/MOX and enriched UO_2 fuel assemblies in a reactor core. A French designed CORAIL assembly was used which employs a standard 17x17 PWR assembly but contains 180 UO_2 fuel rods and 84 MOX fuel rods. In order to maintain reactivity coefficients similar to those in a typical UO_2 fuel assembly, the MOX fuel rods are positioned in the peripheral region of an assembly and the fraction of MOX rods in the assembly is limited to roughly one third of all fuel rod locations [5]. This assembly design was used because even with multiple recycling, the CORAIL assembly has been found to have reactivity coefficients for a full core loading of this assembly type that are comparable to a regular UO_2 fueled core [6].

The deterministic code SCALE (**S**tandardized **C**omputer **A**nalyses for **L**icensing **E**valuation) is a modular code system used to evaluate problem dependent cross-section processing and analysis of criticality safety, and reactor physics problems [7]. First, the SCALE program was used to reproduce the benchmark work already performed by researchers to establish that the quarter of an assembly modeled in SCALE to be used for further investigation could be verified [8]. This was done by using the NEW Transport algorithm for two-dimensional discrete ordinates analysis in non-orthogonal geometries (NEWT) sequence of SCALE to calculate the solution to the two-dimensional transport equation using the 238 ENDF6 energy groups. The dimensions from the CORAIL assembly benchmark were used with a fuel radius of 1.0482 cm, clad radius of 1.2049 cm, and an assembly half pitch of 1.603 cm. Table I shows the fuel composition data for the CORAIL assembly modeled [8].

Table I. Fuel Composition

	ISOTOPE	Atomic Density Atoms/barn-cm
UO2 ROD	U-235	1.1315E-03
	U-238	2.1226E-02
	O-16	4.4716E-02
MOX ROD	U-235	5.2055E-05
	U-238	2.0508E-02
	Pu-238	6.9723E-05
	Pu-239	7.2243E-04
	Pu-240	5.3327E-04
	Pu-241	2.1750E-04
	Pu-242	2.0904E-04
	Am-241	2.1892E-05
	O-16	4.4667E-02

Reactor Core Heterogeneity

An advanced Am/MOX hybrid assembly was modeled in SCALE with the use of two unit cells to represent the two different fuel types in the assembly. The addition of varying amounts of Am-241 in the MOX fuel rods (regular CORAIL composition with 0.03267w/o Am-241, 0.2w/o Am-241, 0.5w/o Am-241, and 1.0w/o Am-241) were used as burnable absorbers. Various branching scenarios, shown in Table II, were incorporated to evaluate the assembly in varying conditions at each burnup step for the purpose of generating functionalized cross-sections for use in core simulations.

Table II. Branching Scenarios

Boron Conc (ppm)	Fuel Temp (K)	Mod Temp (K)	Mod Density (g/cm3)
1300	1600	700	1
950	1300	500	0.85
350	700	400	0.4
50	400	300	0.14

The output from these SCALE calculations was processed for use by NESTLE to fit cross-section data in terms of the foregoing parameters for each of the four assemblies with varying Am-241 concentrations. These libraries include various cross-sections including nu-fission, kappa-fission, transport, absorption, and other data needed to model the different assembly types. NESTLE 5.2.1 was used to model a three dimensional, quarter reactor core with different fuel assemblies with varying Am-241 concentrations. The differences in the eigenvalue (k-inf) for each assembly with varying concentrations of Am-241 can be seen in Table III.

Table III. Am/MOX Hybrid Assembly k-inf

	Fresh Fuel k-inf
MOX	1.31817678
0.2w/oAm/MOX	1.29137688
0.5w/oAm/MOX	1.25289002
1w/oAm/MOX	1.20511605

In order to hold down excess reactivity at the beginning of cycle and to shape the power distribution within the core, the different assemblies were placed in a pattern similar to the traditional use of assemblies with varying amounts of burnable absorbers. The core loading pattern can be seen in Figure 1.

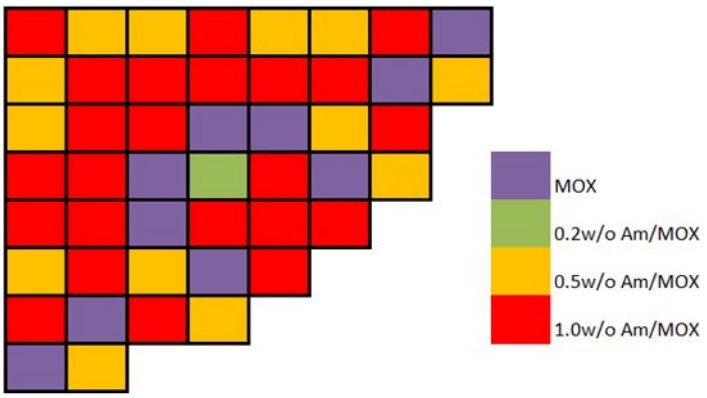

Figure 1. Core Loading Pattern

Because the CORAIL assembly was initially designed to be used in a traditional PWR reactor, a Virgil C. Summer reactor core NESTLE model was used as the basis for this research including the reactor core characteristics, thermal hydraulic data, and the assembly burnup for cycle 17 of the core [1]. The effect of using the advanced Am/MOX assemblies in a PWR was then evaluated with respect to the impact on incineration and transmutation of transuranics as well as the impact on reactor safety parameters.

The selection of assemblies with various Am-241 compositions to be used in this research was based on the k-inf of each assembly as well as core modeling in NESTLE. The substitution of assembly types with varying americium content was made analogous to varying burnable poison loading in the Virgil C. Summer reference core. Following these substitutions, the same core loading pattern was followed. Therefore, as shown in Figure 1, these four fuel assembly types were modeled in the core: MOX, 0.2w/o Am-241 MOX, 0.5w/o Am-241 MOX, and 1.0w/o Am-241 MOX.

RESULTS AND DISCUSSION

With the described core modeled in NESTLE, the criticality of the entire core as a function of burnup was calculated to have a k-eff greater than one throughout the entire cycle (up to 22000 MWD/MTM) and therefore makes this advanced UO_2 and Am/MOX hybrid core viable. To reduce the excess reactivity of the core to the critical condition, boron was used as a chemical shim and varied to compensate for the depletion of the fuel. The effectiveness of the Am/MOX hybrid non-uniform assemblies to hold down the excess reactivity, shape core power distribution, and to transmute the americium was evaluated through a complete suite of analyses.

Radial and Axial Burnup

The initial radial and axial burnup was taken from the assembly exposure during cycle 17 of the Virgil C. Summer reactor core as described in the methodology. This initial exposure included fresh fuel as well as once and twice burned fuel at the beginning of cycle. This initial exposure was read in as input by NESTLE and used in the core calculations. The initial and final exposure of the core averaged axial burnup is lower at the top and bottom of the core as expected due to neutron leakage and the lower relative power at each end of a fuel assembly. Both the radial burnup and core average axial burnup at the end of cycle (Figures 2 and 3) are comparable to

typical PWR assemblies and show that the Am/MOX hybrid non-uniform assemblies can be burned in three cycles, just as with typical UO_2 fuel assemblies.

Figure 2. Radial Burnup at End of Cycle (22000 MWD/MTM)

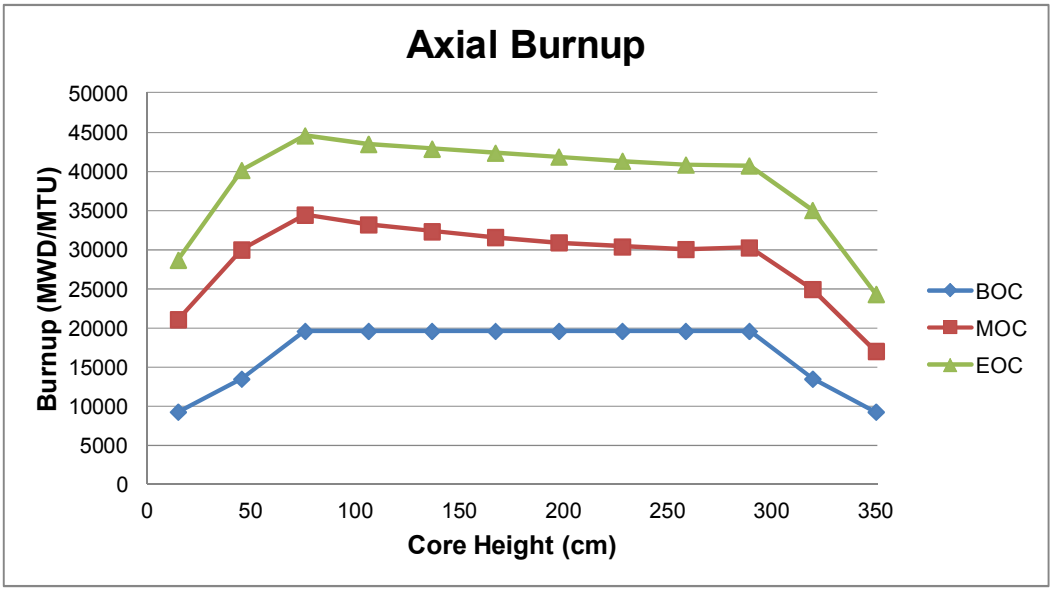

Figure 3. Axial Burnup (MWD/MTM)

Radial and Axial Relative Power

The radial relative power was very significant in this study and was calculated relative to the radial power distribution averaged over the entire core. The same type of Am/MOX hybrid assembly (0.5w/o Am MOX) throughout the entire core was initially tested but yielded a relative maximum power peaking of 1.627 at the beginning of cycle. Therefore, the four different assembly types, each with a different initial reactivity analogous to different assemblies with varying amounts of burnable poisons, were used to reduce this relative power peaking to acceptable levels in a PWR type assembly. The highest assembly relative power peak using the four assembly types was 1.418 at the beginning of cycle, 1.339 at the middle of cycle, and 1.306 at the end of cycle. The radial relative power for each assembly at the beginning, middle, and end of cycle are shown in Figure 4.

1.228	1.088	0.968	0.966	1.237	1.025	0.925	0.289
1.088	1.159	0.993	1.25	1.084	1.216	0.977	0.246
0.967	0.992	1.345	1.418	1.411	1.305	0.563	
0.962	1.243	1.416	1.41	1.393	1.157	0.349	
1.186	1.07	1.401	1.39	1.018	0.445		
1.019	1.207	1.297	1.152	0.444			
0.923	0.973	0.561	0.346				
0.289	0.245						

Beginning of Cycle (0 MWD/MTM)

1.162	0.984	0.945	0.995	1.328	1.153	1.196	0.527
0.984	1.139	0.973	1.218	1.08	1.339	1.178	0.354
0.944	0.973	1.232	1.131	1.174	1.319	0.646	
0.991	1.213	1.13	1.141	1.289	1.143	0.355	
1.284	1.072	1.171	1.288	1.025	0.475		
1.149	1.334	1.317	1.142	0.475			
1.195	1.176	0.645	0.355				
0.527	0.354						

Middle of Cycle (12000 MWD/MTM)

1.306	1.124	1.029	1.013	1.26	1.072	1.109	0.467
1.124	1.252	1.029	1.223	1.046	1.244	1.114	0.38
1.029	1.03	1.25	1.126	1.134	1.246	0.683	
1.011	1.222	1.127	1.116	1.251	1.139	0.423	
1.23	1.041	1.133	1.251	1.073	0.551		
1.07	1.243	1.247	1.14	0.551			
1.109	1.115	0.684	0.423				
0.467	0.38						

End of Cycle (22000 MWD/MTM)

Figure 4. Radial Relative Power at Beginning, Middle, and End of Cycle

In addition to the radial relative power, NESTLE was also used to calculate the axial relative power distribution averaged over the entire core. Not taking into account the initial exposure of the fuel assemblies, the axial relative power is as expected in a PWR type assembly (Figure 5). At the middle of cycle, there is a peak at 110 cm from the bottom of the core which is expected due to the moderator being colder at the bottom of the core and thus providing more moderation and an increased chance of neutron absorption.

However, by including the initial exposure of the once and twice burned fuel assemblies, the axial relative power distribution is significantly different as shown in Figure 6. The axial relative power distribution is bowl shaped at the beginning of cycle because the fresh fuel assemblies with the added americium hold down the most reactivity until some of this americium is transmuted. Therefore, more power initially comes from previously burned fuel that is more depleted in the center of the rod due to higher power over other previous cycles. Once more americium transmutation has occurred in the fresh assemblies, the previously burned assemblies will be holding down the reactivity and thus the middle of cycle relative axial power distribution becomes more like the distribution without the initial exposure.

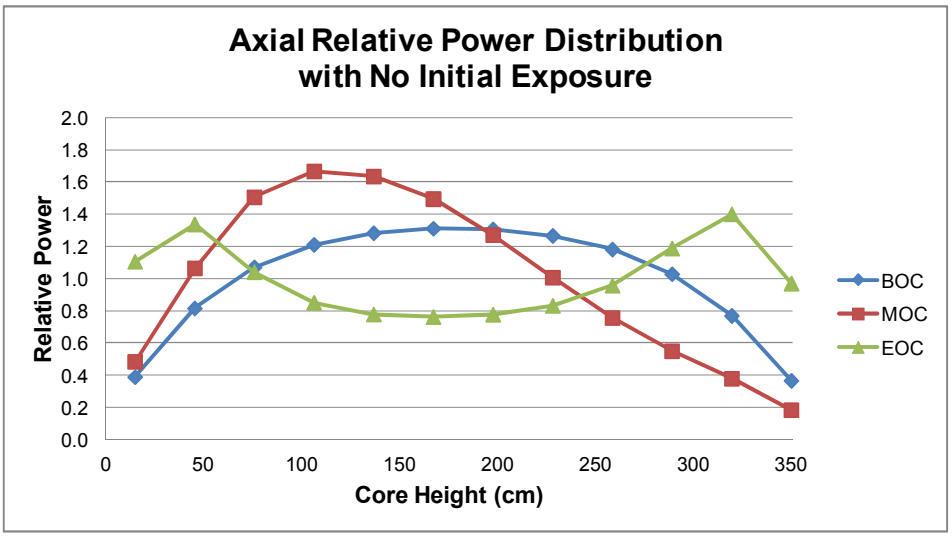

Figure 5. Axial Relative Power Distribution with no Initial Exposure

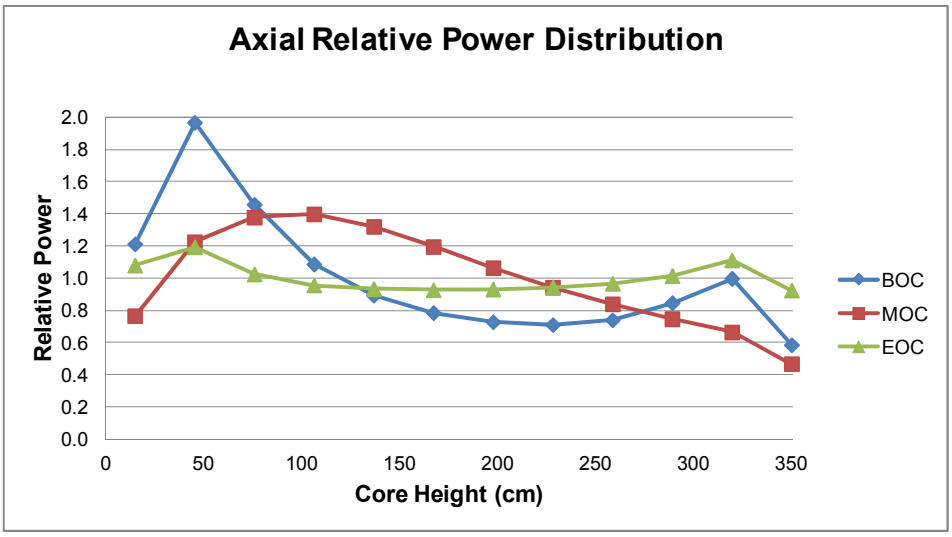

Figure 6. Relative Axial Power Distribution Accounting for Initial Exposure

Power Coefficient of Reactivity

The safety of the americium/MOX hybrid non-uniform core was evaluated through the power coefficient of reactivity. To calculate this coefficient, the power fraction was stepped from almost zero to 100% in 10% increments. NESTLE was used to determine the core criticality at the beginning of cycle for each of these power increments. The reactivity was then calculated at each step based on k-eff. The power coefficient of reactivity was calculated as -0.03565 $\Delta k/k$ at 100% power. The power coefficient of reactivity was sufficiently negative at all power levels to provide protection to the core to prevent an uncontrolled power excursion. The negative power coefficient shows that the core is inherently safe as an increase in power causes a decrease in reactivity.

Temperature and Power Defect

The temperature and power defect was calculated to be -0.03136 and -0.04913 respectively through the evaluation of the cold zero power, hot zero power, and hot full power conditions. Temperature defect is the change in reactivity that occurs in taking the reactor core from fuel loading temperature to zero power operating temperature. The calculated negative temperature defect of -0.03048 is desirable because it leads to core stability as described above. The power defect is the change in reactivity that occurs in taking the reactor core from zero power at operating temperature and pressure to full power. The negative power defect of -0.02767 is due to Doppler cross-section broadening leading to increased resonance absorption. A large jump in power is slowed down by the increased neutron absorption cross-section of the U-238, thus counteracting reactor instability if an uncontrolled power excursion were to occur. All k-eff values were calculated with no xenon and samarium at the beginning of the cycle (0 MWD/MTM). Table IV shows the changes in power, coolant inlet temperature, and coolant pressure for each of these conditions.

Table IV. Reactor Conditions for Temperature and Power Defect Calculation

	Cold Zero Power	Hot Zero Power	Hot Full Power
power fraction	0.0001%	0.0001%	100%
coolant inlet temp (C)	160	284	284
coolant pressure (Pa)	3.45E6	1.55E7	1.55E7
k-eff	1.25914	1.21260	1.17324

Xe-Sm Contribution to Reactivity

Xenon and samarium are important to consider in this reactor core design because they cause significant changes in reactivity due to their large neutron capture cross section and must be accounted for in nuclear reactor analysis. This contribution to reactivity was evaluated through the use of NESTLE by comparing data from Xe-Sm equilibrium option to that of no Xe-Sm option. This was done with no boron added to the moderator and at various steps of the cycle. The xenon and samarium contribution to reactivity for the Am/MOX hybrid reactor core was calculated as -0.02704 at the beginning of cycle and -0.02947 at 22000 MWD/MTU and is comparable to the contribution to reactivity of -0.026 for traditional PWR cores [9].

Control Rod Worth

The control worth of the control rods was evaluated by inserting all eight banks of control rods at various steps and the criticality of the entire core was calculated for each step. The control rods used are of the same type and composition of the Virgil C. Summer reactor core control rods. All k-eff values were calculated with the moderator containing the critical boron concentration, Xe-Sm equilibrium conditions within the core, and at the beginning of the cycle (0 MWD/MTM). The control rod worth at the middle and end of cycle could not be evaluated because the few-group neutron diffusion equation utilizing the nodal expansion method could not be solved by NESTLE beyond several thousand MWD/MTM with the control rods inserted beyond 100 centimeters. The integral control rod worth at a given withdrawal can be seen in Figure 7.

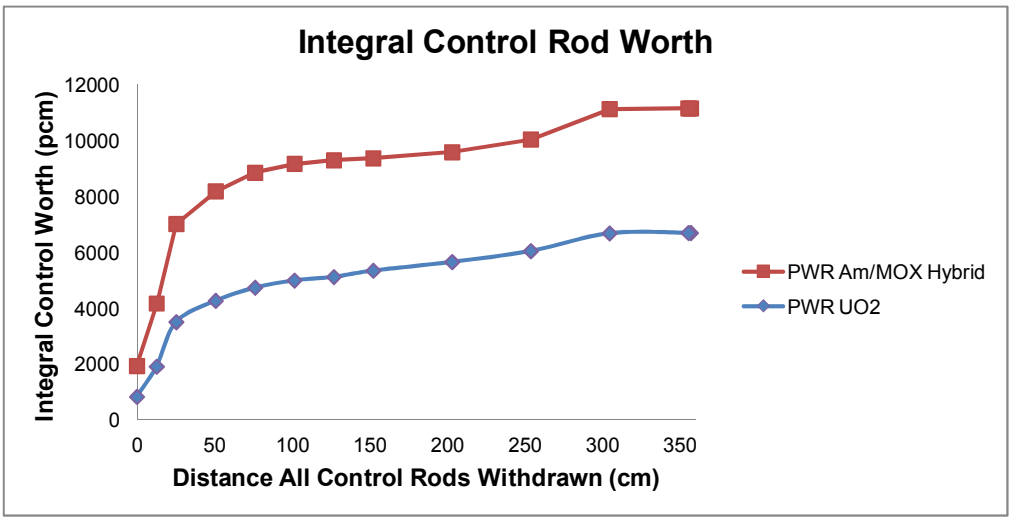

Figure 7. Integral Control Worth at Beginning of Cycle

The use of MOX fuel in an assembly has been shown to degrade the control rod worth because of the large absorption cross-section of Pu-239 in the thermal energy region [10]. One of the main advantages of using the type of non-uniform lattice structures in this analysis over other MOX assembly lattice structures is that this type of assembly only slightly degrades the control rod worth in comparison to a regular PWR assembly containing only UO_2 fuel rods. Because the MOX fuel rods are at the periphery of the assembly with the lattice structure in this research, they are not directly near any control rods, and thus the differential and integral control rod worth is comparable to that of a traditional PWR reactor core.

Transuranic Production and Destruction of Isotopes

SCALE isotopic output of the final composition of 65 metric tons for the entire batch of the Am/MOX hybrid assemblies was used to evaluate the net production and destruction of the transuranic isotopes with the addition of americium to the non-uniform fuel assembly. This included a batch total of 69 assemblies, 16 of which were regular heterogeneous assemblies with no Americium additions to the MOX fuel, 4 of which had a 0.2w/o Am/MOX hybrid, 12 of which had a 0.5w/o Am/MOX hybrid, and 37 of the assemblies had a 1.0w/o Am/MOX hybrid. The isotopic composition of the batch total took into account a core refueling pattern used by traditional PWR reactors in which not every assembly was burned for three cycles.

Accounting for the assemblies that were twice and thrice burned, the elemental composition of the batch at final removal and after a 5 year cooling period is shown in Table V. The percent change shown in the table is calculated as final mass minus initial mass divided by the initial mass. The NA in the table represents an initial concentration so small that a percent change calculation is not useful.

Table V. Batch Fuel Compositions in Grams at Final Removal and After 5 Year Cooling Period

	Fuel Composition (Grams)				
Element	Beginning of Cycle	End of Cycle	(% Change)	After 5 Year Cooling	(% Change)
U	6.682E+07	6.257E+07	-6.36%	6.258E+07	-6.35%
Np	3.074E-11	4.634E+04	NA	4.529E+04	NA
Pu	1.735E+06	2.017E+06	16.30%	1.969E+06	13.52%
Am	4.281E+05	1.833E+05	-57.18%	2.442E+05	-42.96%
Cm	9.456E-11	6.626E+04	NA	4.474E+04	NA
Total	6.899E+07	6.489E+07	-5.94%	6.489E+07	-5.94%

The assembly composition of transuranic isotopes as a function of burnup was also evaluated in this study. Of particular interest in this study is the batch Am-241 composition since this isotope was added to test the transmutation capabilities of this non-uniform lattice (Figure 8).

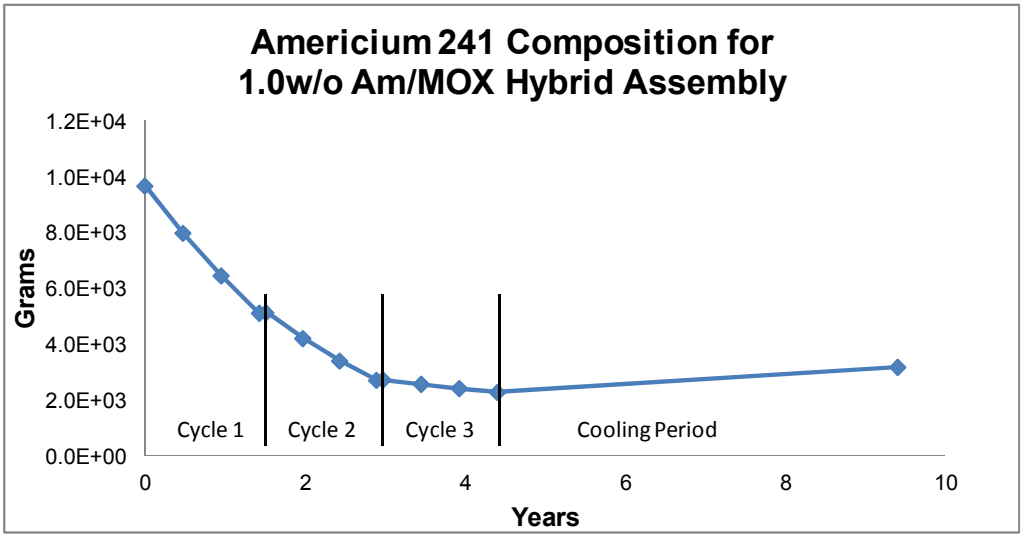

Figure 8. Americium 241 Composition in 1.0w/o Am/MOX Hybrid Assembly

Radiotoxicity

The code ORIGEN-ARP was used to evaluate the long term radiotoxicity impact of the addition of americium to the non-uniform fuel assembly. This code uses a SCALE depletion analysis sequence and an Automatic Rapid Processing (ARP) module that interpolates pre-generated libraries created for a range of isotope properties and then calls ORIGEN-S to compute the time-dependent concentrations and radiation source terms of isotopes, which are simultaneously generated or depleted through neutronic transmutation, fission, and radioactive decay [11]. SCALE isotopic output of the final composition of 65 metric tons for the entire batch of the Am/MOX hybrid assemblies was used as the fuel composition to be decayed by ORIGEN, taking into account a core refueling pattern of twice and thrice burned assemblies. Figure 9 shows the activity in Curies of the batch waste after final removal from the core as the isotopes decay as a function of time in years.

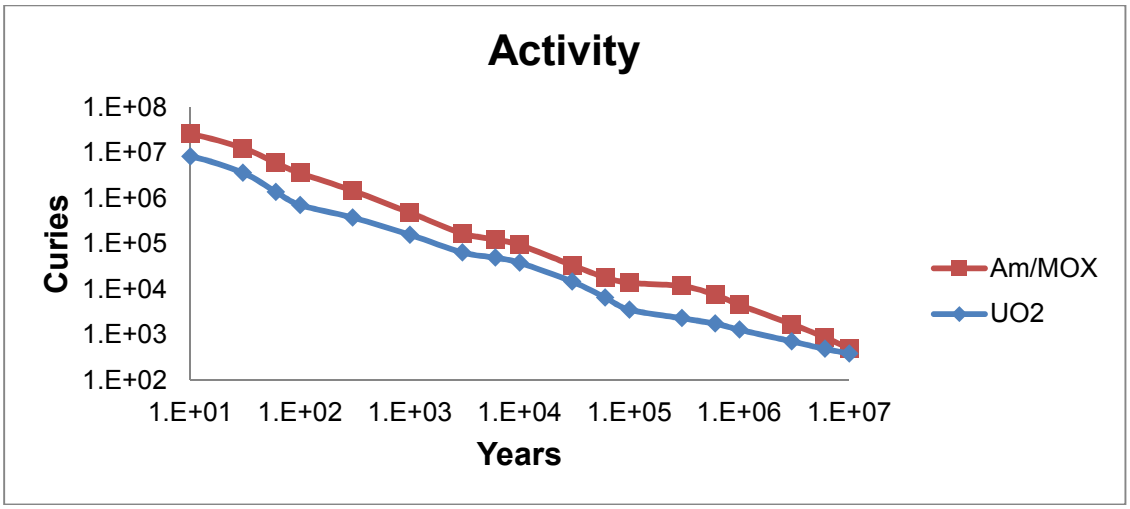

Figure 9. Activity of a Single Batch Waste in Curies

In order to calculate the radiotoxicity of the depleted material, the cancer risk coefficients for environmental exposure to radionuclides from the Environmental Protection Agency's Federal Guidance Report 13 were used. The risk coefficients are the risk per Becquerel of mortality from ingestion averaged over all ages and genders. To obtain Sv/Bq, this risk coefficient was then divided by the age averaged cancer mortality risk estimate per Sievert from a low dose uniform irradiation of the body from the report [12]. These factors were applied to the activity from the output of ORGIN and summed for all nuclides in the waste material, thus expressing the radiotoxicity in Sievert.

The radiotoxicity was then normalized to the amount of energy produced by the assemblies per TWHe. As part of the normalization calculation, a batch average burnup of 55000 MWD/MTM was assumed. Also accounted for in this normalization were the TRU isotopes used to produce the various Am/MOX hybrid assemblies. In order to produce the plutonium required for each Am/MOX hybrid assembly, 10 regular UO_2 assemblies must be used (assuming a burnup of 55000MWD/MTU). In order to produce the Am-241 added to a 0.2w/o Am/MOX assembly, 50 regular UO_2 assemblies must be used and a 0.5w/o Am/MOX assembly and a 1.0w/o Am/MOX assembly require 124 and 247 regular UO_2 assemblies respectively. Uranium and plutonium were assumed to be recycled and thus not used in the radiotoxicity calculation. Figure 10 shows the radiotoxicity normalized using the method described.

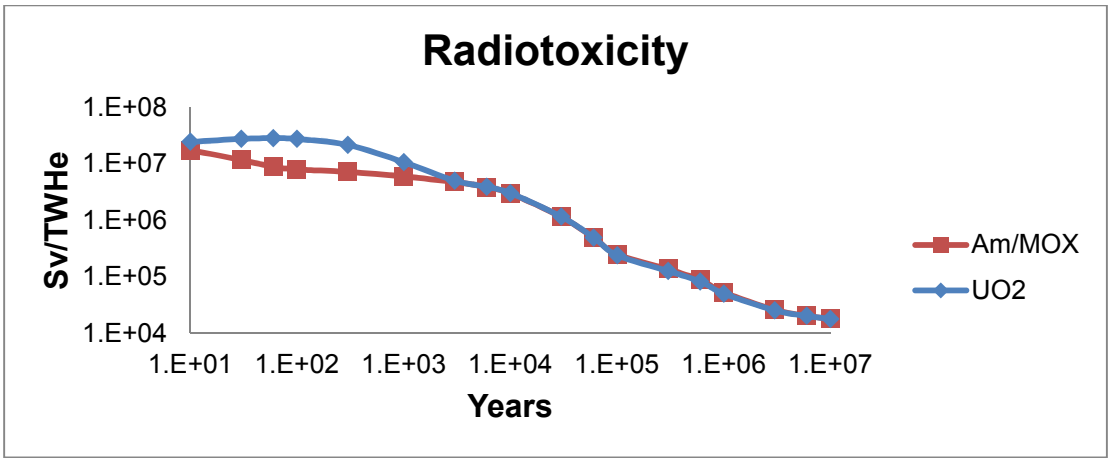

Figure 10. Radiotoxicity of Single Batch Waste in Sieverts Normalized to 1 TWHe

The radiotoxicity of the high level waste that must be managed from a once used Am/MOX hybrid batch is less than a regular UO_2 batch for the first 1000 years until most of Am-241 in the UO_2 batch waste decays to Np-237 through alpha particle emission. While this radiotoxicity analysis does take into account the regular UO_2 assemblies that must be used in the production of the Am/MOX hybrid assemblies, it does not factor in multiple recycling. As shown through numerous studies, the heterogeneous assembly model used as the basis for this research is capable of multiple recycles while still maintaining negative temperature and power coefficients of reactivity.

Heatload

ORGIN was also used to evaluate the impact on heatload of the high level waste material by adding the americium to the non-uniform MOX assemblies. The heatload in watts was normalized using the same methodology described in the radiotoxicity analysis by taking into account the TRU isotopes used to produce the various Am/MOX hybrid assemblies as well as the amount of energy produced by the assemblies per TWHe. There is no advantage in heatload of the Am/MOX hybrid non-uniform assembly batch high level waste without multi-recycling as compared to regular UO_2 batch high level waste. However, the use of multi-recycling should reduce this heatload to less than that of a once through UO_2 fuel cycle.

CONCLUSION

The use of Am/MOX hybrid fuel assemblies to transmute americium was researched using multiple computer codes. Am-241 was shown in this study to be able to hold down excess reactivity at the beginning of cycle and shape the power distribution in the core with assemblies of varying americium content loaded in a pattern similar to the traditional use of assemblies with varying amounts of burnable absorbers.

The feasibility, safety, and utility of using americium to create an Am/MOX hybrid non-uniform core were also evaluated. The core remained critical to a burnup of 22,000 MWD/MTM. The power coefficient of reactivity as well as the temperature and power defects were sufficiently negative to provide a prompt feedback mechanism in case of a transient and prevent a power excursion, thus ensuring inherent safety and protection of the core. As shown here as well as many other studies, this non-uniform assembly type successfully addresses the concerns of

reduced control rod worth within advanced MOX assemblies because the Am/MOX hybrid fuel rods are on the periphery of the assembly. The radiotoxicity of the high level waste that must be managed from a single use of an Am/MOX hybrid batch is reduced for the first 1000 years in comparison to a regular UO_2 batch. However, there is no heatload advantage without multi-recycling the Am/MOX hybrid assemblies.

As shown through numerous studies, the heterogeneous assembly model used as the basis for this research is capable of multiple recycles while still maintaining negative temperature and power coefficients of reactivity. Multi-recycling would provide an even greater reduction to the radiotoxicity of the high level waste as well as provide a heatload advantage compared to a once through UO_2 fuel cycle. This research concludes that the addition of americium to a non-uniform fuel assembly is a viable method of transmuting Am-241, holding down excess reactivity in the core while serving as a burnable poison, as well as reducing the radiotoxicity of high level waste that must be managed.

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