

Transmutation of Plutonium and Minor Actinide in PWR Thorium-Transuranic Fuel Assembly

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Abstract—Long-lived radioactive waste is often considered a concerning issue on utilizing nuclear power. This waste is in the form of plutonium and minor actinides (MA), formed due to successive neutron capture of ^{238}U . Both are not particularly hazardous radiologically, but their long half-life caused an issue in the public acceptance of radioactive waste disposal. Thereby, this issue must be resolved, either politically or technically. One of the technical solutions to address the issue of long-lived radioactive waste is the incineration of transuranic (TRU) elements in a pressurized water reactor (PWR) fuel assembly. Mixing TRU with thorium in a PWR fuel assembly can theoretically reduce TRU stockpile more effectively than uranium. This paper discusses plutonium and MA transmutation in a Thorium-Transuranic (Th-TRU) PWR fuel assembly using MCNP6 code and ENDF/B-VII library. A Westinghouse 17×17 PWR fuel assembly was chosen to determine the feasibility of TRU incineration. Three assemblies of 50:50, 55:45, and 60:40 mixture of UO₂ and Th-TRU fuel rods are compared to one reference UO₂ assembly from the criticality and burn-up point of view. From the calculations, the plutonium incineration rate was observed to be 9.47 %, 10.91 %, and 12.2 %, while MA incineration rates were found to be 11.5 %, 11.93 %, and 12.3 % for Th50-TRU50, Th55-TRU45, and Th60-TRU40, respectively. This observation indicates that a higher thorium fraction in the fuel assembly can increase the TRU transmutation rate. Therefore, a high thorium fraction is recommended to increase the transmutation rate of the TRU.

Keywords—Transmutation; plutonium and minor actinide; Th-TRU; PWR fuel assembly.

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I. INTRODUCTION

Although the risk is exaggerated, long-lived radioactive waste is often considered concerning in utilizing nuclear power. The waste in concern is plutonium and minor actinides (MA), formed due to successive neutron capture of ^{238}U . Both are not particularly hazardous radiologically, but their long half-life caused an issue in the public acceptance of radioactive waste disposal. Therefore, this issue must be resolved, either politically or technically. One of the technical solutions to focus on the issue of long-lived radwaste is the incineration of transuranic (TRU) elements in a pressurized water reactor (PWR) fuel assembly. Mixing TRU with thorium in a PWR fuel assembly can theoretically reduce TRU stockpile more effectively than uranium.

PWR constitutes the large majority of the operating nuclear power plants. The reactor is light water-cooled and fueled with low-enriched uranium in the form of uranium dioxide (UO₂) pellets stacked within a Zircaloy-clad fuel rod.

PWR technology has been well established, but its fuel cycle has generated a constant issue throughout its operational history. Leftover transuranic (TRU) elements from discharged PWR spent fuel are somewhat troublesome in terms of long-lived high-level radioactive waste issues.

The TRU consists of plutonium (Pu) and minor actinide (MA) isotopes. The Pu is highly degraded and comprises more than 20 % of ^{240}Pu ; therefore, it is unsuitable for proliferation attempts. Meanwhile, MA mainly consists of curium (Cm), americium (Am), and neptunium (Np). While Np and ^{241}Am have particular usefulness, other MA isotopes are generally useless and simply add to the burden of long-lived radwaste disposal. Transmuting TRU is the most logical technical solution to reduce the burden of radioactive waste disposal. As PWR is the most mature nuclear power reactor technology, it can potentially be used as a medium for TRU transmutation to reduce its stockpile. However, simply recycling TRU into the uranium fuel cycle does not seem beneficial. Recycling TRU into the thorium fuel cycle can

theoretically be a better alternative due to the lower atomic mass of thorium, providing a significantly longer path to TRU generation.

Thorium (Th) is a vastly untapped nuclear fuel, and its utilization potential is hindered by the absence of its natural fissile isotope. Introducing TRU which has a fissile isotope (^{239}Pu) into Th, can help its utilization along with incinerating the TRU. The usefulness of Th application in PWRs has been confirmed in several publications. The feasibility study of ThMOX fuel utilization and its behavior in light water reactors was discussed [1], [2] while exploring the possible advantages of using thorium-based fuel in a PWR was analyzed in a total core calculation [3]. The optimization of Th utilization was also performed in a Reduced Moderation PWR (RMPWR) instead of a regular PWR. The former has a harder spectrum that works better for thorium [4]. The neutronic analysis of thorium MOX fuel in a prismatic high-temperature gas-cooled reactor [5] and a comparative analysis of the neutronic performance of thorium mixed with uranium or plutonium in a high-temperature pebble-bed reactor [6] was conducted. However, none of the above publications discussed the incineration of TRU within a homogeneous Th-TRU fuel. Analysis of MA incineration in Russian PWR was performed, but not within a Th-TRU fuel [7].

This research aims to explain some results concerning the transmutation of Pu and MA in a Th-TRU fuel assembly using MCNP6 radiation transport code [8] and continuous energy nuclear data library ENDF/B-VII [9]. A Westinghouse 17×17 fuel assembly [10] is the representative model. In this work, the TRU is mixed with Th as a homogeneous fuel rod and placed within UO_2 fuel assembly in a distributed configuration. Attention is focused on the Th-TRU fuel assemblies' criticality and burn-up characteristics to determine the relationship between Th fraction and TRU incineration.

II. MATERIALS AND METHOD

A. PWR Fuel Assembly

The PWR fuel assembly has a square array of 17×17 rods consisting of 264 fuel rods, an instrumentation tube, and 24 guide tubes. Guide tubes are cylindrical metal tubes used as structural support to the fuel assembly, a conduit for inserting control rods, and channels for burnable absorber rods if used. The instrumentation tube, which is generally a central guide tube, provides structural support for the fuel assembly as well as channels for the insertion of in-core monitoring instrumentation. Guide tubes and instrumentation tubes are typically made from Zircaloy. The design of the Westinghouse fuel assembly and fuel rod that is used in this work are provided in Tables 1 and 2.

The fuel assembly is cooled and moderated with pressurized light water at 0.723 g/cm³ density. For the general assembly, the UO_2 fuel is enriched to 4.5 wt % of ^{235}U with a density of 10.41 g/cm³. The fuel pellet itself has a diameter of 0.805 cm, with a fuel rod pitch of 1.26 cm and a fuel assembly height of 365 cm. Zircaloy-4 cladding encased each fuel rod and tube within the assembly with a thickness of 0.0571 cm. In total, the mass of uranium in the fuel assembly is 450,030 grams. The fuel is then irradiated at a

constant power of 54 MWt for 360 days, resulting in a discharge burn-up of 43 GWd/MTU.

TABLE I
PWR FUEL ASSEMBLY

Fuel assembly type	Westinghouse 17×17 PWR
Fuel assembly pitch	21.42 cm
Reactor operating power	54 MW
Number of fuel rods per assembly	264
Total fuel burn-up	43 MWd/kgHM
Total of guide tubes per assembly	24
Total of instrumentation tubes per assembly	1
Reactor operating time	360 days

TABLE II
PWR FUEL ROD

Fuel pellet	
Fuel material	UO_2
Fuel density	10.41 g/cm ³
Diameter of fuel	0.805 cm
Fuel height	365 cm
Temperature of fuel	900 K
Rod pitch of fuel	1.26 cm
$^{234}\text{U}/^{235}\text{U}/^{238}\text{U}$ initial mass	180.2/20,250/429,600 g
Fuel enrichment	4.5 % ^{235}U
Total uranium mass	450,030 g
Cladding	
Cladding material	Zircaloy-4
Density of cladding	6.52 g/cm ³
Thickness of cladding	0.0571 cm
Temperature of cladding	622 K
Moderator	
Moderator/coolant material	H_2O
Moderator density	0.723 g/cm ³
Temperature of moderator	576 K
Concentration of Boron	850 ppm

TABLE III
FOUR DIFFERENT TYPE OF FUEL ASSEMBLY

Fuel assembly	Number of fuel rod	Th-TRU fuel content
Th0-TRU0	264 UO_2 , 0 Th-TRU	-
Th50-TRU50	200 UO_2 , 64 Th-TRU	50 % thorium, 50 % TRU
Th55-TRU45	184 UO_2 , 80 Th-TRU	55 % thorium, 45 % TRU
Th60-TRU40	158 UO_2 , 96 Th-TRU	60 % thorium, 40 % TRU

This work considered three Th-TRU assemblies and one UO_2 reference assembly. The Th-TRU assemblies comprise different Th:TRU weight ratios, which are 50:50, 55:45, and 60:40. Each is designated as Th50-TRU50, Th55-TRU45, and Th60-TRU40, respectively. Meanwhile, the reference assembly is designated as Th0-TRU0. While the reference Th0-TRU0 assembly is fully loaded by 264 UO_2 rods, other fuel assemblies have the UO_2 rods mixed with a different number of Th-TRU fuel rods with each designated thorium fraction. The Th-TRU rods are placed in a distributed configuration, meaning that the rods are placed in a specific distribution pattern within the assembly. The number of Th-TRU fuel rods for each Th-TRU assembly is given in Table 3. Different numbers of fuel rods are set in such a way so that the initial infinite multiplication factor (k_{inf}) of each fuel

assembly is not too dissimilar to the reference UO_2 assembly in order to provide a fair start.

B. Calculation Model

A series of numerical computations using the MCNP6 code and ENDF/B-VII neutron cross-section library has been performed to present the transmutation of Pu and MA in a Th-TRU PWR fuel assembly. MCNP6 is a general-purpose Monte Carlo radiation transport software that has been demonstrated to be sufficiently accurate in simulating many reactor physics parameters in various nuclear reactors [11]–[27]. The code can precisely model complicated geometries and track the evolution of temporal nuclide inventory by simulating the actual physical process with continuous energy coefficients. MCNP6 supports the depletion capability using CINDER90 module. CINDER90 is a burn-up and depletion code that uses Markovian chain to solve the sets of time-dependent Bateman equations. It calculates isotope depletion and new number densities of isotopic formation of fuel materials using the destruction and creation coefficients and normalization constant provided by MCNP [28], [29].

The MCNP6 model of detailed PWR fuel assembly was done by first modeling fuel rod, guide and instrumentation tubes in a cubic lattice with a pitch of 1.26 cm. The fuel rod consists of 0.4025 cm-radius fuel pellets surrounded by a 0.0571 cm thick Zircaloy-4 cladding. Water with 850 ppm boron concentration was used as coolant and moderator at the outside of the fuel rod. The guide and instrumentation tubes have the same inner and outer radius of 0.4025 cm and 0.4496

cm, respectively, even though they are used for different purposes. The model of these tubes is the same as the fuel rod model, but where there are supposed to be fuel pellets, they are replaced with coolant water. The PWR fuel assembly is then modeled by creating an array of 17×17 rods consisting of fuel rods, guide, and instrumentation tubes. The lattice models of fuel rods and tubes are shown in Fig. 1. Four different models of fuel assembly comprised of UO_2 and Th-TRU fuel rod structures are laid out in Fig. 2.

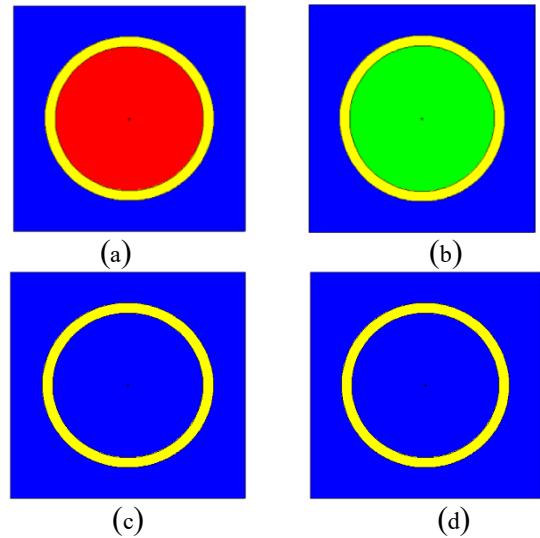


Fig. 1 MCNP6 model of fuel rod and tubes: a – UO_2 ; b – Th-TRU; c – Guide tube; d – Instrumentation tube

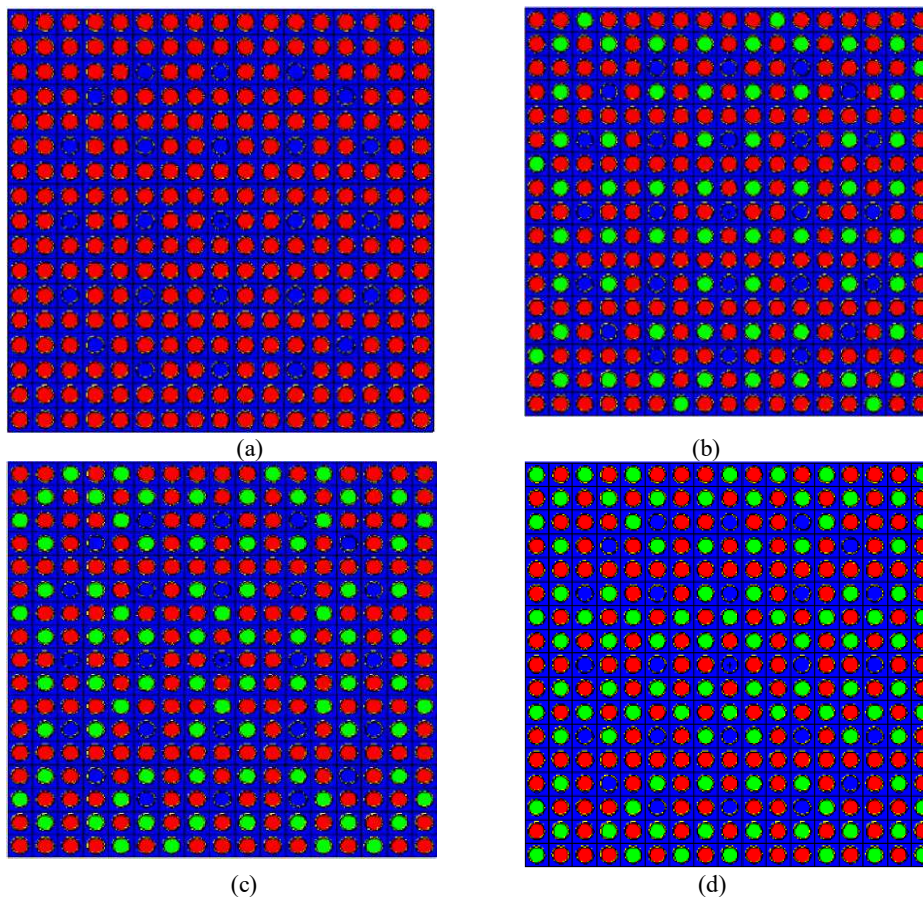


Fig. 2 MCNP6 model of Pressurized Water Reactor fuel assembly: a – Th0-TRU0; b – Th50-TRU50; c – Th55-TRU45; d – Th60-TRU40

TABLE IV
REACTOR GRADE PLUTONIUM VECTOR (%)

^{238}Pu	1.58	^{241}Pu	8.76
^{239}Pu	57.76	^{242}Pu	5.33
^{240}Pu	26.57		

TABLE V
MINOR ACTINIDES VECTOR (%)

^{237}Np	42.25	^{243}Cm	0.01
^{241}Am	47.57	^{244}Cm	1.26
^{243}Am	8.51	^{245}Cm	0.07
^{242}Cm	0.32	^{246}Cm	0.01

The isotopic composition of the reactor-grade Pu and MA vectors, as given in Tables 4 and 5, are derived from a 3000 MWt PWR spent fuel composition with a burn-up of 33 GWd/THM after ten years of cooling. The Pu and MA mass ratio in the spent fuel is 1:9 [30]. TRU and Th are mixed within a single oxide fuel in the form of (Th,TRU) O_2 . The assumed density of Th O_2 is 9.5 g/cm 3 , while that of TRU O_2 is 10.36 g/cm 3 .

The MCNP6 calculation determined the temperatures of coolant, fuel, and clad materials at 600 K, 900 K, and 600 K, respectively. Neutron transport within the fuel assembly was simulated with 10,000 neutron histories per cycle and 300 total cycles consisting of 250 active cycles and 50 first cycles discarded. The initial neutron sources were distributed at several locations in the assembly. The thermal scattering law $S(\alpha, \beta)$ of Iwtr.26t (600K) was applied for the treatment of the binding effect of hydrogen on H $_2\text{O}$ to its interaction with neutrons at thermal energies. BURN card was used to employ CINDER90 to perform fuel burn-up calculations. For all simulations, the reflective boundary conditions were applied to all fuel assembly sides.

III. RESULTS AND DISCUSSION

The k_{inf} calculation results are shown in Fig. 3. As previously mentioned, the number of Th-TRU fuel rods in Th-TRU assemblies is different from maintain the k_{inf} value as close as possible to the reference assembly. Consequently, TRU loading within the three assemblies is also different. Higher thorium fraction requires more fissile loading to provide a comparable initial k_{inf} value. This results in Th60-TRU40 having the largest TRU loading in the assembly, despite in its individual rods, TRU loading is the lowest.

From Fig. 3, it can be observed that k_{inf} evolution in Th-TRU assemblies is not significantly different to each other. k_{inf} of Th60-TRU40 assembly declined faster in the first few days, but beyond that, its k_{inf} declined slower than Th55-TRU45 and Th50-TRU50. At day 360, Th60-TRU40 has the highest k_{inf} , albeit the difference is insignificant. The high Th loading in the assembly causes such pattern. As more Th captured neutrons, k_{inf} decreased more drastically at the beginning of irradiation. However, after ^{233}U formed from Th, k_{inf} increased because it neutronically outperforms ^{235}U and ^{239}Pu in thermal spectrum. High Th loading in Th60-TRU40 helped the ^{233}U formation to be higher, although the effect would only appear in the later stage of burn-up. Meanwhile, reference UO $_2$ assembly has its k_{inf} dropped

most drastically, implying that Th-TRU addition can indeed increase the fuel cycle length without needing higher excess reactivity.

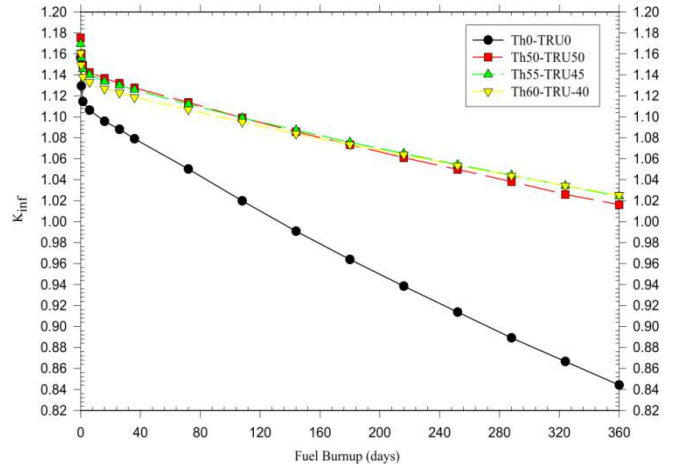


Fig. 3 k_{inf} as a function of burn-up

The transmutation of selected Pu isotopes for all assemblies is shown in Figs. 4–6. Pu concentration will undoubtedly increase in reference assembly as a transmutation result of fertile ^{238}U . Meanwhile, in Th-TRU assemblies, the only isotope to show a declining concentration is ^{239}Pu . This is understandable as ^{239}Pu is the driver of fission reaction in the Th-TRU assemblies. The increase of ^{240}Pu and ^{242}Pu are a direct consequence of neutron capture of fissile ^{239}Pu and ^{241}Pu , when the captured neutron did not trigger a fission reaction. Around 20 % of the neutron capture of ^{239}Pu and ^{241}Pu caused transmutation into a higher number of isotopes instead of fission.

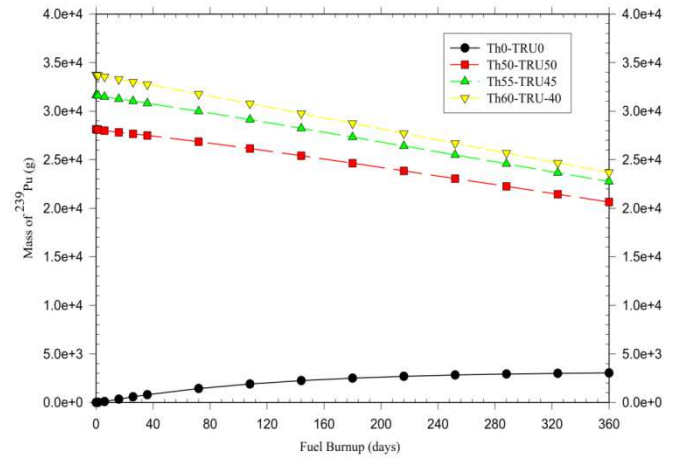


Fig. 4 Transmutation of ^{239}Pu as a function of burn-up

Nevertheless, ^{240}Pu increase is comparably lower than ^{242}Pu . The former has considerably high neutron capture cross section, transmuting it into fissile ^{241}Pu . The rate of ^{240}Pu formation from ^{239}Pu capture is only slightly higher than ^{240}Pu transmutation into ^{241}Pu . This is an indication that ^{240}Pu acted like a neutron poison.

The Pu transmutation rate is summarized in Table 6 as a mass comparison at the beginning of cycle (BOC) and at the end of cycle (EOC). As discussed above, the only isotope which mass is decreasing is ^{239}Pu as the primary driver of

fission reaction in Th-TRU fuel rods. Other Pu isotopes have their mass increasing, including fissile ^{241}Pu which fission rate is lower than its formation rate from ^{240}Pu . However, for each assembly, the transmutation rate is different. Th60-TRU40 assembly has the highest transmutation rate; around 7,145 g (12.2 %) of Pu is destroyed mostly by fission reaction.

Meanwhile, Th55-TRU45 transmutation rate is 5,990 g (10.9 %), and Th50-TRU50 is 4,630 g (9.5 %). Although the total remaining Pu at EOC is the highest, a transmutation in Th60-TRU40 is more effective. This is due to the transmutation phenomenon calculated within the individual fuel rods. A higher Th fraction ensures that the TRU is transmuted more efficiently, so when the transmutation in individual Th-TRU rods is summed up, the result is a larger Pu reduction. This is helped by the fact that Th-TRU fuel rods are in a distributed configuration, ensuring that the individual rods receive higher neutron flux than in a seed-and-blanket configuration.

The mass fraction of ^{239}Pu in all Th-TRU assemblies is lowered from the initial $\pm 57\%$ to $\pm 46\%$. The initial reactor grade Pu is already degraded so much that the attempt for proliferation is futile. Lowering its mass fraction even more while simultaneously increasing ^{240}Pu fraction renders the last bit of possible attempt for proliferation completely useless. All this while providing ample usable energy in the nuclear reactor and initiating the formation of ^{233}U , which is important in the thorium fuel cycle.

For MA transmutation, the selected results are displayed in Figs. 7–10. Similar to the Pu case, reference UO₂ assembly has its MA isotopes concentration increase, although the ^{243}Am and ^{244}Cm increase are less apparent before day 200. In all Th-TRU assemblies, only ^{237}Np and ^{241}Am have the largest fraction, showing mass decrease, with the latter showing a more drastic decrease. Apart from high mass fraction, both isotopes have comparably higher thermal capture cross sections than other MA isotopes listed in Table

7, causing their macroscopic cross sections to be the largest and, consequently, easiest to capture neutrons and then transmute [31]. Meanwhile, other MA isotopes show a constantly increasing trend because their macroscopic capture cross section is insufficient to take over the dominant capture reaction from their preceding MA isotopes.

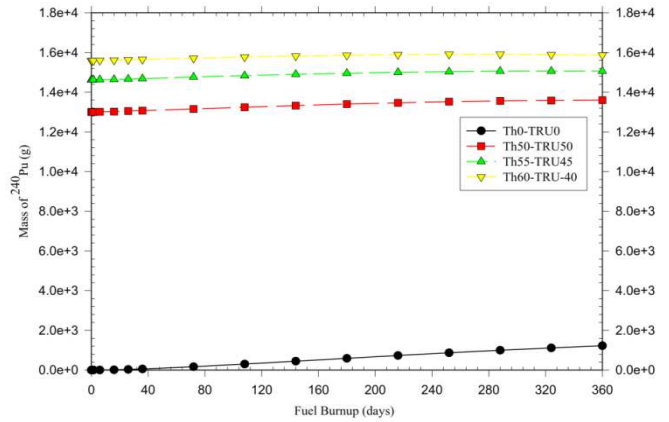


Fig. 5 Transmutation of ^{240}Pu as a function of burn-up

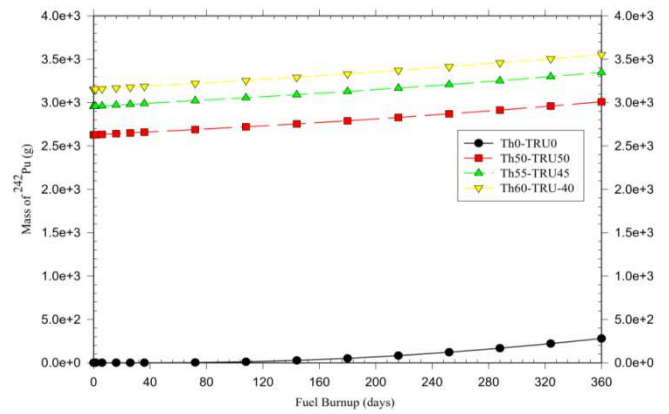


Fig. 6 Transmutation of ^{242}Pu as a function of burn-up

TABLE VI
TRANSMUTATION OF PLUTONIUM ISOTOPE

Th50-TRU50 fuel assembly			
Isotope	Pu mass at 0 day (g)	Pu mass at 360 days (g)	Transmutation rate (%)
^{238}Pu	766.3 (1.6 %)	1,699 (3.8 %)	-121.7
^{239}Pu	28,130 (57.6 %)	20,640 (46.7 %)	26.6
^{240}Pu	13,000 (26.6 %)	13,600 (30.8 %)	-4.6
^{241}Pu	4,302 (8.8 %)	5,253 (11.9 %)	-22.1
^{242}Pu	2,629 (5.4 %)	3,009 (6.8 %)	-14.5
Total	48,830	44,200	9.5
Th55-TRU45 fuel assembly			
Isotope	Pu mass at 0 day (g)	Pu mass at 360 days (g)	Transmutation rate (%)
^{238}Pu	862.0 (1.6 %)	1,906 (3.9 %)	-121.1
^{239}Pu	31,640 (57.6 %)	22,750 (46.5 %)	28.1
^{240}Pu	14,620 (26.6 %)	15,060 (30.8 %)	-3.0
^{241}Pu	4,839 (8.8 %)	5,863 (12.0 %)	-21.2
^{242}Pu	2,957 (5.4 %)	3,348 (6.8 %)	-13.2
Total	54,920	48,930	10.9
Th60-TRU40 fuel assembly			
Isotope	Pu mass at 0 day (g)	Pu mass at 360 days (g)	Transmutation rate (%)
^{238}Pu	919.2 (1.6 %)	2,046 (4.0 %)	-122.6 %
^{239}Pu	33,750 (57.6 %)	23,690 (46.1 %)	29.8 %
^{240}Pu	15,590 (26.6 %)	15,870 (30.9 %)	-1.8 %
^{241}Pu	5,161 (8.8 %)	6,270 (12.2 %)	-21.5 %
^{242}Pu	3,153 (5.4 %)	3,552 (6.9 %)	-12.7 %
Total	58,573	51,428	12.2 %

TABLE VII
TRANSMUTATION OF MINOR ACTINIDE ISOTOPE

Th50-TRU50 fuel assembly			
Isotope	MA mass at 0 day (g)	MA mass at 360 days (g)	Transmutation rate (%)
^{237}Np	2,267 (41.8 %)	1,567 (33.9 %)	30.9
^{241}Am	2,596 (47.9 %)	1,561 (33.8 %)	39.9
^{243}Am	468.3 (8.6 %)	652.0 (14.1 %)	-39.2
^{242}Cm	17.54 (0.3 %)	431.2 (9.3 %)	-2358.4
^{243}Cm	0.550 (0.01 %)	20.52 (0.4 %)	-3628.9
^{244}Cm	69.62 (1.3 %)	349.7 (7.6 %)	-402.3
^{245}Cm	3.884 (0.07 %)	39.53 (0.9 %)	-917.8
^{246}Cm	0.557 (0.01 %)	2.079 (0.05 %)	-273.2
Total	5,423	4,800	11.5

Th55-TRU45 fuel assembly			
Isotope	MA mass at 0 day (g)	MA mass at 360 days (g)	Transmutation rate (%)
^{237}Np	2,550 (41.8 %)	1,902 (35.4 %)	25.4
^{241}Am	2,920 (47.9 %)	1,763 (32.8 %)	39.6
^{243}Am	52.67 (8.6 %)	750.8 (14.0 %)	-42.5
^{242}Cm	19.72 (0.3 %)	485.5 (9.0 %)	-2362.0
^{243}Cm	0.619 (0.01 %)	23.27 (0.4 %)	-3659.9
^{244}Cm	78.31 (1.3 %)	399.5 (7.4 %)	-410.2
^{245}Cm	4.368 (0.07 %)	45.87 (0.9 %)	-950.1
^{246}Cm	0.627 (0.01 %)	2.348 (0.04 %)	-274.7
Total	6,100	5,372	11.9

Th60-TRU40 fuel assembly			
Isotope	MA mass at 0 day (g)	MA mass at 360 days (g)	Transmutation rate (%)
^{237}Np	2,720 (41.8 %)	1,994 (34.9 %)	26.7
^{241}Am	3,114 (47.9 %)	1,855 (32.5 %)	40.4
^{243}Am	561.7 (8.6 %)	812.6 (14.2 %)	-44.7
^{242}Cm	21.03 (0.3 %)	526.2 (9.2 %)	-2402.1
^{243}Cm	0.660 (0.01 %)	25.45 (0.4 %)	-3755.5
^{244}Cm	83.51 (1.3 %)	438.5 (7.7 %)	-425.1
^{245}Cm	4.658 (0.07 %)	51.26 (0.9 %)	-1000.5
^{246}Cm	0.668 (0.01 %)	2.601 (0.05 %)	-289.3
Total	6,506	5,706	12.3

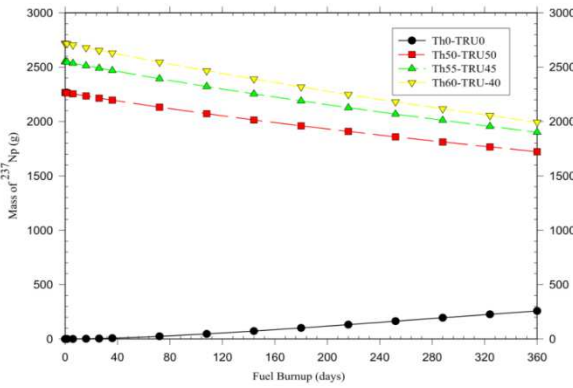


Fig. 7 Transmutation of ^{237}Np as a function of burn-up

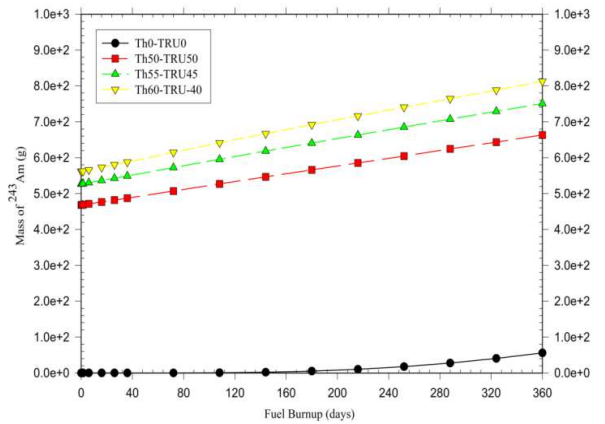


Fig. 9 Transmutation of ^{243}Am as a function of burn-up

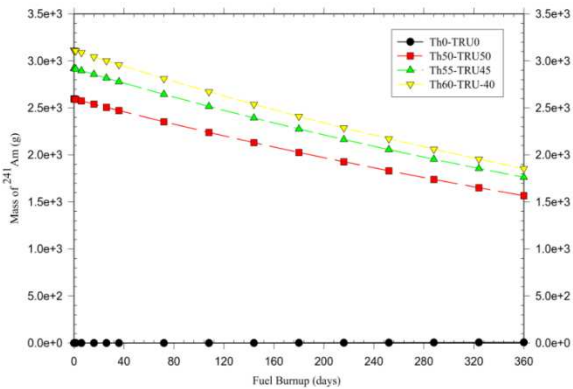


Fig. 8 Transmutation of ^{241}Am as a function of burn-up

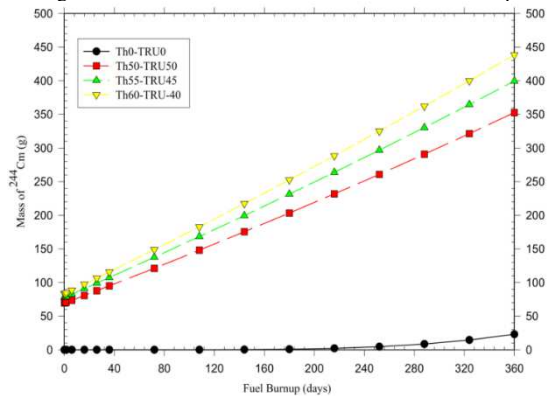


Fig. 10 Transmutation of ^{244}Cm as a function of burn-up

As the case with Pu, the Th60-TRU40 assembly also shows the largest MA transmutation rate, decreasing its mass by 800 g (12.3 %) compared to Th55-TRU45 of 728 g (11.9 %) and Th50-TRU50 of 623 g (11.5 %). MA transmutation rate for all cases is higher than Pu due to MA being parasitic to neutrons. The incineration rate is lower than the result obtained by Tran et al. [7]. However, this work mixed the MA in Th-TRU fuel rods instead of using it as a burnable poison, so the difference in transmutation rate is understandable.

IV. CONCLUSION

The transmutation of plutonium and minor actinide in the fuel assembly of PWR has been conducted using MCNP6 radiation transport code and ENDF/B-VII nuclear data library. In the Th-TRU assemblies, the attainable Pu transmutation after 360 days of irradiation is 9.5 %, 10.9 %, and 12.2 %, while for MA are 11.5 %, 11.9 %, and 12.3 % for Th50-TRU50, Th55-TRU45, and Th60-TRU40 assemblies, respectively. The increase of Th fraction coupled with a larger number of Th-TRU fuel rods in the fuel assembly can improve the effective reduction of Pu and MA mass. This paper did not explore the effect of Th-TRU rod addition in PWR fuel assembly in a distributed configuration towards the reactor physics and safety characteristics, such as power distribution, delayed neutron fraction, Doppler reactivity, and coolant void temperature coefficient. Those aspects must be investigated in the forthcoming works.

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