

INVESTIGATION ON NEUTRONIC BEHAVIOR OF PEBBLE BED REACTOR FOR TRU TRANSMUTATION

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Abstract. One alternative effort to eliminate the accumulation of transuranic (TRU) elements generated from LWR spent fuel is by utilizing TRU as nuclear fuel in pebble bed reactor. This work was aimed to investigate the neutronic behavior of a pebble bed reactor used for TRU transmutation. The reactor geometry is adopted from the HTR-Modul. MCNP6 multipurpose radiation transport code with ENDF/B-VII.1 neutron library was used to calculate the neutronic aspects. The results indicated that the initial effective multiplication factor (k_{eff}) values decrease as the amount of TRU fuel pebbles within the core increases, concurrent with the fuel burnup. Increasing the TRU fuel pebble ratio resulted in weakened Doppler temperature coefficients (DTCs) and improvement in moderator temperature coefficients (MTCs). The calculated total mass of plutonium and minor actinide transmutation demonstrated that a pebble bed reactor with 100% TRU pebble can reduce TRU mass up to 55.47% of its initial mass load.

Key words: neutronic behavior, pebble bed reactor, TRU transmutation, MCNP6, ENDF/B-VII.1.

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1. INTRODUCTION

Pebble bed reactor, a variant of Very High Temperature Reactor (VHTR), has been placed as the first priority for the Generation IV reactor to be built. Its inherent safety feature is characterized by helium coolant, graphite moderator with a ceramic core, and TRISO-coated fuel particles. Helium is non-reactive, thus do not react to form explosive gas or underwent phase change. The core structure and moderator consist of graphite, which have high thermal conductivity. The ceramic, multi-layered fuel particle allows the integrity of fission product containment under the design temperature limit of 1600°C [1].

The accumulation of transuranic elements (TRU) generated from spent fuel of Light Water Reactor (LWR) is particularly challenging due to potential material

diversion, high radiotoxicity, and long half-life [2]. TRU primarily consists of plutonium (Pu) isotopes and smaller number of minor actinides (*i.e.* Np, Am, and Cm). TRUs are formed *via* successive neutron capture of actinides with higher atomic number than uranium ($Z = 92$). After discharged, TRU comprises around 1% of the discharged spent fuel. Their properties impact the nuclear fuel cycle, mostly as determinant of requirements and procedures regarding the back-end of fuel cycle [3].

Among the possible efforts to overcome the accumulation of TRU is by utilizing TRU as nuclear fuel in thermal and fast reactors. Several fast reactor core design concepts were carried out for TRU transmutation such as the PEACER lead alloy cooled fast reactor with metallic fuel (U, TRU) 10%Zr designed in South Korea to perform TRU transmutation [4], the KALIMER core designed by KAERI for future SFR applications to transmute TRU [5]. AREVA and MHI develop the Advanced Recycling Reactor (ARR) by providing the examination of a more efficient TRU-burning fast reactor core [6]. The characteristics of TRU recycling on the shifted neutron spectrum within a Pb-Bi core was carefully examined by considering two Pb-Bi cooled cores, namely soft and hard spectrum [7]. The Prototype Gen-IV Sodium-cooled Fast Reactor (PGSFR) was designed as a TRU transmutation reactor by applying several oxide fuel-loaded TRU [8].

Fast reactors are the mostly considered for transmuting TRU as fast neutron energy is generally more effective in obliterating TRU. However, studies have demonstrated that TRU can also be transmuted in thermal neutron energy, although with severe limitations due to the TRU accumulation through recycling and possibly negative impact on the safety of the plant. A considerable amount of research has been conducted on TRU transmutation in thermal reactors, such as AP1000 with some selected fuels including PuO_2 , Pu_3Si_2 , PuN, and MOX, etc. in a multi-pin assembly [9], PWR with Th-TRU fuel which has demonstrated design feasibility for TRU annihilation despite having lower control rod worths and smaller delayed neutron fraction [10]. The TRU transmutation in Combined Non-Fertile and UO_2 (CONFU) PWR assembly was evaluated consisting of a mixture of regular UO_2 fuel pins and TRU pins in an inert matrix and designed to be applicable in currently operating PWRs [11].

Pebble bed reactor is another thermal reactor that has the flexibility to offer the possibility of operating in a full TRU-fueled core to incinerate plutonium and transmute minor actinides without significant change to core structure design. However, the TRU fueled core is predicted to have a less favorable neutronic behavior given the high concentration of plutonium (Pu) content in TRU fuel. This work is aimed to investigate the neutronic behavior of pebble bed reactor for TRU transmutation. The TRU was taken from PWR spent fuel with a burnup of 45 GWD/MTHM after 30-years of cooling. The HTR-Modul [12] was chosen as the reactor model. It has been investigated extensively as a reference design for the South African Pebble Bed Modular Reactor (PBMR) project [13] and HTR programs in China [14].

Neutronic calculations were performed using MCNP6 Monte Carlo code [15] and ENDF/B-VII.1 neutron cross section library [16]. The fuel burnup calculations were deployed by utilizing the CINDER90 [17] module integrated in the MCNP6 code. MCNP6 is a multi-purpose Monte Carlo radiation transport code and has been demonstrated extensively in the simulation of core physics parameters for various types of nuclear reactors [18–27]. In this study, the core configuration consisting of combination of TRU and UO₂ fuel pebbles with five composition ratios were simulated and the results were compared to each other to analyze their neutronic performances in TRU transmutation.

2. PEBBLE BED REACTOR

The German concept of HTR-Modul [28] is a cylindrical core with a diameter and a height of 3 m and 9.43 m, respectively. The reactor uses helium as the coolant fluid and graphite as moderator as well as structural material. The reactor thermal power is 200 MW with power density of 3 MW/m³. The HTR-Modul was designed on the criterion that no active safety protection system is required due to its inherently safe nature. The reactor was projected to generate electricity and industrial process heat.

Table 1

Design parameter of HTR-Modul [28]

Reactor parameter	
Thermal power (MW)	200
Helium inlet/outlet temperature (°C)	250/750
Helium pressure (MPa)	6
Helium mass flow rate (kg/s)	85
Helium coolant density (kg/cm ³)	4.33×10^{-3}
Core specification	
Core power density (MW/m ³)	3
Diameter (cm)/height (cm)	300/943
Number of fuel pebble per m ³	5,394
Number of fuel pebble in core	359,548
Pebble packing fraction in core	0.61

The reactor core is surrounded by ceramic structures consisting of graphite reflectors. A core cavity made of a void with a height of 50 cm is located above the fuel region. The shutdown system is provided in the graphite reflector to control the core reactivity and shutting down the reactor in all conditions. Helium enters the reactor core at a temperature of 250°C, then flows down through the gaps between the pebbles while extracting heat, and carries the heated gas at a temperature of

750°C to the turbine to generate electricity. Helium is recirculated back to the reactor through channels within the graphite reflector. The design parameter of HTR-Modul is given in Table 1.

A total of 359,548 fuel pebbles are randomly arranged within the core with a pebble packing fraction of 0.61. The fuel pebbles are loaded into the top of the core and discharged from the bottom. HTR-Modul adopts a multi-pass refueling strategy. In this strategy, a fuel handling system is prepared to undertake the mechanisms for fuel discharging, burnup measuring, and reloading into the core. The fuel pebble can be recirculated up to ten times if it does not reach the specified burnup. If the fuel pebble has attained the target burnup, it will then be discharged and transferred into spent fuel cask for storage and disposal.

Each fuel pebble has a diameter of 6 cm, consisting of 15,000 TRISO-coated randomly-dispersed fuel particles within a graphite matrix in 5 cm-diameter central zone. The central zone is surrounded by a 0.5 cm thick graphite shell. A fuel particle consists of a 0.050 cm-diameter UO_2 kernel wrapped with a buffer layer, inner PyC layer, SiC layer, and outer PyC layer, consecutively, with the thicknesses of 90 μm , 40 μm , 35 μm , and 40 μm , respectively. The UO_2 kernel has a ^{235}U enrichment of 8.2%. The packing fraction of the TRISO particles is 9.043% [29].

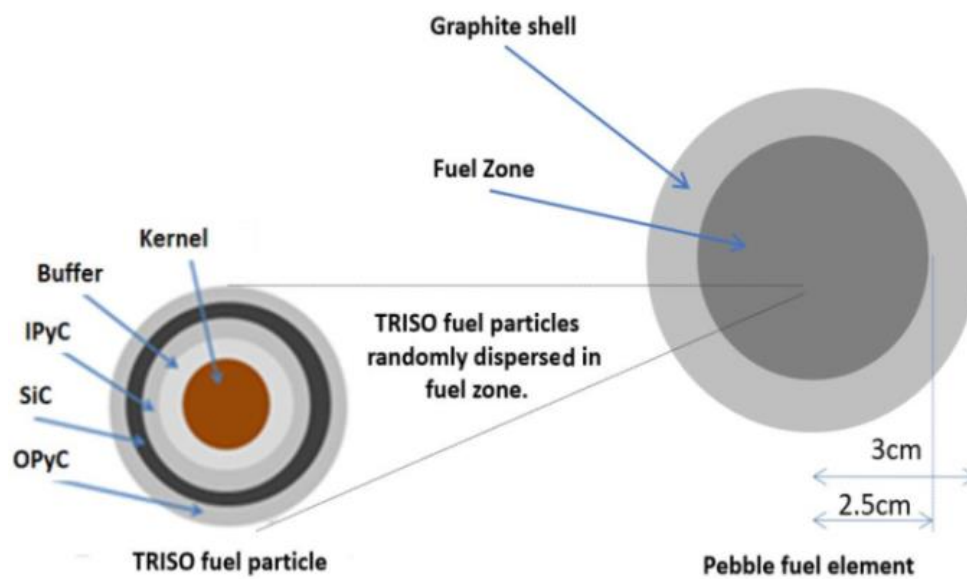


Fig. 1 – The schematic view of fuel pebble [30].

Meanwhile, TRU kernel of 24 μm in diameter is wrapped by the same coating layers with a different thickness in the buffer layer of 95 μm . The packing

fraction of coated particle with TRU kernel is 3.45%, its smaller packing compared to UO_2 kernel is because its overall TRISO radius is smaller. In general, the TRISO layers act as multiple barriers to prevent the leak of fission products from the fuel particle and ensuring that particle failure do not occur even at extremely high burnup for a wide range of operating conditions below the 1600°C temperature limit.

The fuel pebble model is depicted in Fig. 1, with specifications of UO_2 and TRU fuel pebble given in Table 2. The UO_2 density is 10.40 g/cm^3 which corresponds to a uranium content of 10.21 g per fuel pebble. In TRU fuel, the isotopic vector is produced from a PWR spent fuel after 30 years of cooling with a discharge burnup of 45 MWD/kgHM [31] as given in Table 3. The ^{239}Pu fraction in the TRU vector is quite high with a total fissile Pu of around 59%. The TRU used is in the form of TRU- O_2 with a density of 10.36 g/cm^3 . The transuranic content in the TRU fuel pebble is 0.9927 g.

Table 2

Specification of fuel pebble

Fuel pebble		
Fuel composition	UO_2 , TRU O_2	
Fuel pebble radius (cm)	3	
Fueled zone radius (cm)	2.5	
Graphite density in matrix/shell (g/cm^3)	1.75	
Natural boron impurity in fuel/ graphite (ppm)	1.0/0.5	
TRISO-coated particle		
Material	Density (g/cm^3)	Thickness (cm)
Kernel	10.40 ^a	0.0500 (dia) ^b
Buffer	1.05	0.0090 ^c
iPyC	1.90	0.0040
SiC	3.18	0.0035
oPyC	1.90	0.0040

^a10.36 for TRU, ^b0.0240 for TRU, ^c0.0095 for TRU fuel

Table 3

Isotopic TRU vector (%)

^{237}Np	4.69
^{238}Pu	1.27
^{239}Pu	56.27
^{240}Pu	20.11
^{241}Pu	3.04
^{242}Pu	3.08
^{241}Am	9.99
^{243}Am	0.77
^{244}Cm	0.06

3. CALCULATION MODEL

Modeling pebble bed reactor is different with LWR due to the existence of double heterogeneity, comprised of irregularly located TRISO-coated particles in the pebble fuel and similarly irregular pebbles location in the core. The solution to the double heterogeneity problem is essential to get accurate results for the design, safety analysis, and operation of the reactor. MCNP6 can solve the double heterogeneity problem both in irregular and regular ways. In this study, the irregular TRISO particle locations in the fuel pebble and the irregular location of the pebble fuel in the core were approached as repeated structures.

Model of Fuel Pebble. Prior to fuel pebble modeling, the TRISO particle was modeled with a simple cubic (SC) cell. The real geometry and exact dimensions of the TRISO particle composed of the kernel and all four coating layers were used. The SC unit cell with a length of 0.16343 cm was defined as a UNIVERSE and constructed by a few cells based on the surface. After that, the 15,000-unit cells of fuel particles were filled in the pebble with a regular way to complete the model of the fuel pebble. The repetitive structure capability of LATTICE was implemented in the fuel pebble modeling. The use of a repetitive structure option will produce truncated TRISO particles by the boundary of the fueled zone in the pebble. However, its effect can be ignored because the calculations in regular and irregular ways do not affect the results significantly [32].

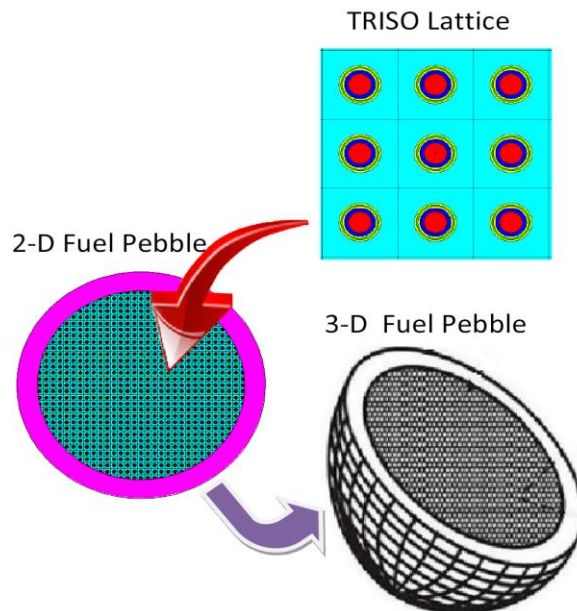


Fig. 2 – Model of fuel pebble.

The fuel pebble model is illustrated in Fig. 2. Nuclide concentration of TRISO particles with UO₂ and TRU kernels is provided in Table 4 while concentrations for the graphite matrix and graphite shell are summarized in Table 5.

Table 4

Nuclide concentration of TRISO particle

Fuel kernel UO ₂ (atom/barn-cm)			
²³⁵ U	1.92585×10^{-3}	¹⁶ O	4.64272×10^{-2}
²³⁸ U	2.12877×10^{-2}	¹⁰ B	1.14694×10^{-7}
¹¹ B	4.64570×10^{-7}		
Fuel kernel TRU (atom/barn-cm)			
²³⁷ Np	1.10981×10^{-3}	²⁴² Pu	7.20767×10^{-4}
²³⁸ Pu	3.14433×10^{-4}	²⁴¹ Am	2.28088×10^{-3}
²³⁹ Pu	1.29758×10^{-2}	²⁴³ Am	1.94733×10^{-4}
²⁴⁰ Pu	4.63126×10^{-3}	²⁴⁴ Cm	3.38257×10^{-5}
²⁴¹ Pu	7.14630×10^{-4}	¹⁶ O	4.59523×10^{-2}
Coating layers (atom/barn-cm)			
Porous carbon buffer		iPyC/oPyC	
¹² C	5.26449×10^{-2}	¹² C	9.52621×10^{-2}
SiC			
¹² C	4.77240×10^{-2}	²⁹ Si	2.22871×10^{-3}
²⁸ Si	4.40158×10^{-2}	³⁰ Si	1.47944×10^{-3}

Table 5

Nuclide concentration of TRISO particle

Graphite matrix (atom/barn-cm)		Graphite shell (atom/barn-cm)	
¹² C	8.77414×10^{-2}	¹² C	8.77414×10^{-2}
¹⁰ B	9.64977×10^{-9}	¹⁰ B	9.64977×10^{-9}
¹¹ B	3.90864×10^{-8}	¹¹ B	3.90864×10^{-8}

Model of Reactor Core. In order to model the reactor core, a face-centered cubic (FCC) cell of fuel pebble was constructed. The FCC cell is formed by four pebbles consisting of 8 pebbles of 1/8 at the corners and 6 pebbles of 1/2 at each cube faces. The length of a unit cell is 9.05286 cm which was found from the relation between the in-core pebble packing fraction (0.61) and the volume of four pebbles inside the unit cell. The pebble bed core was modeled by filling 359,548 fuel pebbles into the core in a regular way. The repetitive structure capability of LATTICE was implemented again in the modeling of the reactor core.

Pebbles intersected with the core surface, as a result of the use of repetitive structures, should be corrected because it will add extra fuel to the core and indirectly influence the accuracy of the calculation results. The correction was made by creating a 3 cm thick exclusive zone of helium around the reactor core.

This methodology was first proposed by Lebenhaft [33] with excellent results and utilized as a reference to model the geometry of pebble bed reactors in various publications [34-42]. The reactor core model is depicted in Fig. 3.

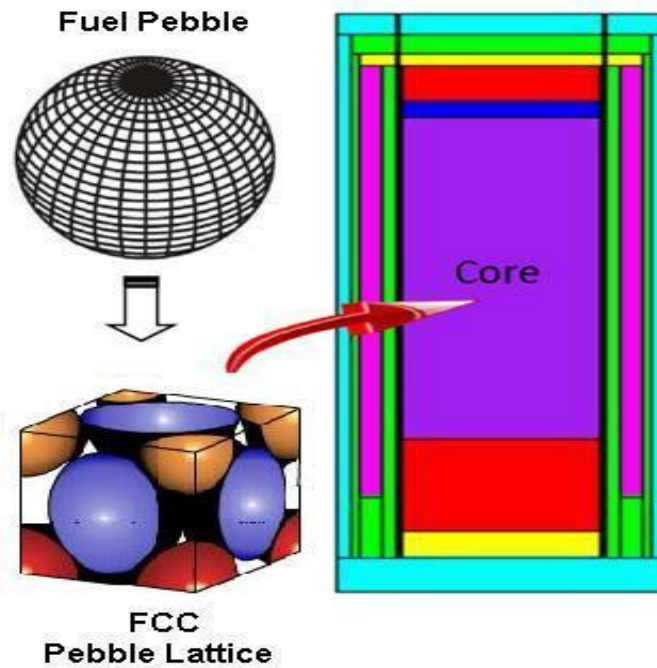


Fig. 3 – Model of reactor core.

In this study, the core configuration consists of a combination of TRU and UO_2 fuel pebbles with five ratio compositions. Based on four pebbles in the FCC unit cell, the composition of TRU and UO_2 fuel pebbles can be arranged in core configuration with ratios of 0%/100%, 25%/75%, 50%/50%, 75%/25%, and 100%/0%, referred as to TRU0, TRU25, TRU50, TRU75, and TRU100, respectively. For instance, TRU25 is a core configuration with a composition ratio of 25% TRU and 75% UO_2 fuel pebbles constructed by $8 \times (1/8)$ TRU pebbles at the corners and $6 \times (1/2)$ UO_2 pebbles at the faces of the unit cell.

In the MCNP6 calculation, 10,000 neutron histories per iteration and 250 active iterations were used with 50 initial iterations discarded to converge the distribution of fission source. The initial fission sources were located at the central part of the fuel kernel. The thermal scattering data $S(\alpha, \beta)$ for graphite was applied to take into account the interaction of thermal neutron with carbon at the energy region below ~ 4 eV. CINDER90 depletion module was utilized for fuel burnup calculations. In the kinetics parameters calculation, the KOPTS card was activated and the temperatures of fuel, moderator, and other regions were set to 900 K.

4. RESULTS AND DISCUSSION

The effective multiplication factor (k_{eff}) and temperature coefficient of reactivity from combined TRU and UO_2 fuel pebbles are presented in Table 6. k_{eff} is a nuclear reactor parameter related to fission reaction sustainability to maintain a neutron population in the reactor core. As shown in Table 6, k_{eff} values decrease as the share of TRU fuel pebbles in the core increases. This is primarily related to the absorption of ^{240}Pu that is greater than ^{238}U . ^{240}Pu is a fertile isotope that dominates the TRU fuel, followed by ^{237}Np .

Table 6

Effective multiplication factor and temperature coefficient of reactivity ($\Delta k/k/K$)

Configuration	Effective multiplication factor (k_{eff})	Doppler temperature coefficient (DTC)	Moderator temperature coefficient (MTC)
TRU0	1.30703 ± 0.00055	-4.70232×10^{-5}	-2.67352×10^{-5}
TRU25	1.21590 ± 0.00052	-4.10073×10^{-5}	-5.47575×10^{-5}
TRU50	1.17089 ± 0.00053	-3.35142×10^{-5}	-5.97481×10^{-5}
TRU75	1.14725 ± 0.00049	-2.47336×10^{-5}	-5.81428×10^{-5}
TRU100	1.13572 ± 0.00047	-1.49693×10^{-5}	-5.46820×10^{-5}

Temperature coefficient of reactivity, which characterizes the fuel behavior during increasing temperature, could assess the inherent safety feature of nuclear fuel. Two most important reactivity coefficients are considered, namely Doppler temperature coefficient (DTC) and moderator temperature coefficient (MTC). DTC relates to the contribution of the Doppler broadening on the nuclide reaction cross-section, which could change the absorption-to-fission ratio of fuel material. MTC is closely related to the moderator-to-fuel ratio that can affect neutron moderation, which then change the reactivity under the change of graphite temperature.

DTC was determined by changing the temperature of the fuel from 293 K to 1200 K while the temperature of the moderator and other regions are kept the same. Table 6 confirms that all the DTCs are negative with increasing TRU fuel pebble ratio giving a smaller DTC value, which is attributable to the smaller Doppler effect on capture cross-section by ^{238}U isotope from fewer UO_2 fuel pebble in the reactor core. These results show that using TRU fuels could reduce DTC.

Total reactivity feedback in the reactor core is a combined effect from DTC and MTC, the latter of which was determined by varying the moderator temperature from 293 K to 1200 K while preserving the fuel temperature and other regions. Shown in Table 6, TRU-mixed cores have the MTC increased and reached the maximum of -5.97481×10^{-5} at 50% and slightly decreased at 75% till 100%. The negative value on MTC shows that the core is in over-moderated conditions, otherwise, the effective multiplication factor decrease when TRU pebbles increase

showing that existing neutron moderation could not improve thermal neutron fission reaction with this new fuel mixture. These fewer thermal neutrons to induce fission caused the MTC to become more negative. Combined with DTC, MTC provide additional inherent safety in case of increasing temperature during reactor operations.

Table 7

Kinetic parameters

Configuration	Effective delayed neutron fraction (β_{eff})	Prompt neutron lifetime (ℓ , sec)	Neutron generation time (Λ , sec)
TRU0	0.00524 ± 0.00050	$1.1937 \times 10^{-3} \pm 1.3987 \times 10^{-6}$	$6.6430 \times 10^{-4} \pm 8.1198 \times 10^{-6}$
TRU25	0.00451 ± 0.00050	$9.6973 \times 10^{-4} \pm 1.4361 \times 10^{-6}$	$5.7852 \times 10^{-4} \pm 6.9688 \times 10^{-6}$
TRU50	0.00348 ± 0.00040	$8.6837 \times 10^{-4} \pm 1.3280 \times 10^{-6}$	$5.1053 \times 10^{-4} \pm 6.3931 \times 10^{-6}$
TRU75	0.00280 ± 0.00030	$8.0495 \times 10^{-4} \pm 1.2791 \times 10^{-6}$	$4.7299 \times 10^{-4} \pm 6.3509 \times 10^{-6}$
TRU100	0.00247 ± 0.00032	$7.6771 \times 10^{-4} \pm 1.2623 \times 10^{-6}$	$4.4371 \times 10^{-4} \pm 6.3576 \times 10^{-6}$

The results of kinetic parameters calculation are presented in Table 7. The parameters consisting of effective delayed neutron fraction (β_{eff}), prompt neutron lifetime (ℓ), and neutron generation time (Λ) indicate the ability to control a reactor and reactivity response due to power perturbations. It can be observed that the β_{eff} values decrease with the increased number of TRU pebble. The β_{eff} value decreases by more than 50% when the UO_2 fuel pebble is completely replaced by TRU. These results signify the β_{eff} is highly dependent on fissile material within fuel pebble which in TRU, with ^{239}Pu has a lower β_{eff} value than ^{235}U . A lower β_{eff} value means that the core will have a faster response to perturbation and tends to deliver a faster transient behavior that needs to be controlled. In terms of safety features, a higher β_{eff} value gives a relatively slow response for a reactor to a perturbation that could give additional time to control. But in general, the difference is minimum since there are many other parameters that could give additional responses, *i.e.* control rod, and coolant flow rate that affect temperature distribution within the reactor core.

The same thing occurs to ℓ and Λ , which tends to become smaller when the TRU fuel pebble fraction increases. It is well known that increasing TRU fuel fraction will shift the neutron spectrum to become harder. With large neutron capture cross-section in TRU isotopes and as their fraction increases within the core, it will decrease ℓ and Λ , because these parameters are inversely proportional to the core average macroscopic absorption cross-section [43].

The effect of a higher TRU fuel pebble fraction on the reactivity coefficient and kinetic parameters is a determinant of the neutronic characteristics of the reactor core. This could lead to a change in neutron population and its safety parameter when compared to uranium-fueled cores which are generally used in pebble bed reactors.

The evolution of k_{eff} throughout burnup is depicted in Fig. 4. The initial k_{eff} for the core fully loaded with UO_2 fuel pebble (TRU0 core) is the highest, but it only maintains k_{eff} value above unity up to 70 MWd/kgHM. Combinations of TRU and UO_2 fuel pebbles could increase this maximum burnup value, such as TRU25, TRU50, and TRU75 core configurations can achieve fuel burnups of 90, 100, and 200 MWd/kgHM, respectively. Pebble bed core loaded with TRU fuel only (TRU100 core) was observed to have a lowest initial k_{eff} value, which decreases slowly and reaches 500 MWd/kgHM until it becomes subcritical (below unity). The more TRU fuel pebble added to the core, the higher its achieved maximum burnup. This is due to the presence of several fissile materials on TRU fuel pebbles such as ^{239}Pu and ^{241}Pu and fertile material that could transmute into fissile, such as ^{238}Pu , ^{240}Pu , ^{241}Am , ^{243}Am , and ^{244}Cm . This combination provides a longer operating time for TRU fueled core with lower initial excess reactivity.

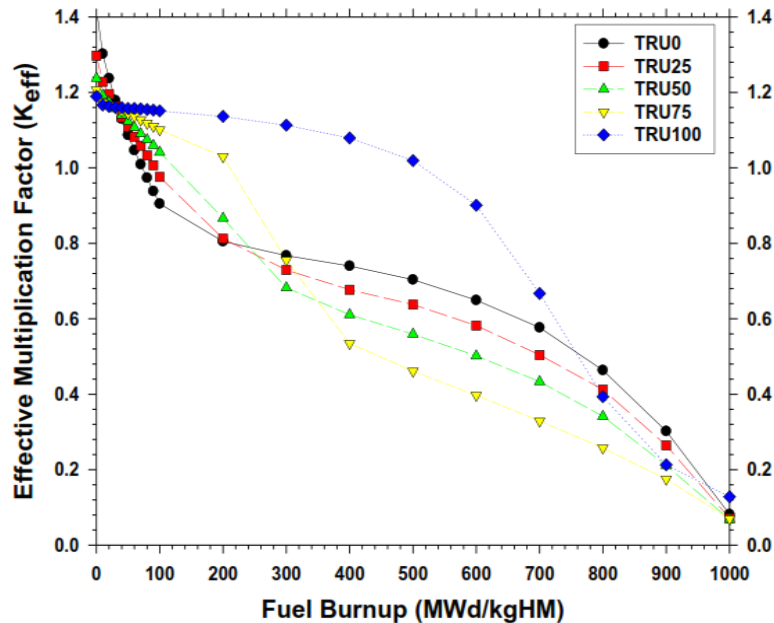


Fig. 4 – K_{eff} as a function of burnup.

Such condition will be advantageous in terms of excess reactivity which must be suppressed at the beginning of cycle. In regard to safety, with lower excess reactivity and flatter reactivity change, the core dependence on the control rod could be reduced. By maintaining proper fuel management, this type of core could operate safely without changing the control rod position, which could lead to replacing its function to become a shutdown rod. In this case, the control rod only focuses on shutting down the reactor if unexpected conditions occur. By adjusting

the number of TRU fuel pebble fractions, the core can be critical in longer time which could lead to fewer downtime.

Plutonium Transmutation. Figures 5–9 show plutonium isotopes concentration as a function of fuel burnup, ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu , respectively. As predicted, plutonium concentration is higher in a core with TRU fuel pebble compared to those that only use UO_2 fuel pebble. In general, it can be seen from Fig. 6 that the ^{239}Pu concentration is the highest compared to other Pu isotopes because the initial amount of ^{239}Pu is quite large in TRU fuel pebble. In UO_2 fuel pebble, ^{238}U will capture neutrons and undergo several beta decays to become ^{239}Pu . Furthermore, by capturing neutrons, ^{239}Pu can transmute to other heavier isotopes.

The amount of Pu isotopes was varying due to the accumulation of several isotopes that undergo (n,2n) and (n,3n) reactions, and some of them are also fissile *i.e.* ^{239}Pu and ^{241}Pu . Some of ^{239}Pu and ^{240}Pu isotopes will also undergo neutron capture and transmuted into ^{242}Pu . It made the peak concentration of plutonium isotope at the end of burnup occur not only at ^{238}Pu but also at ^{242}Pu .

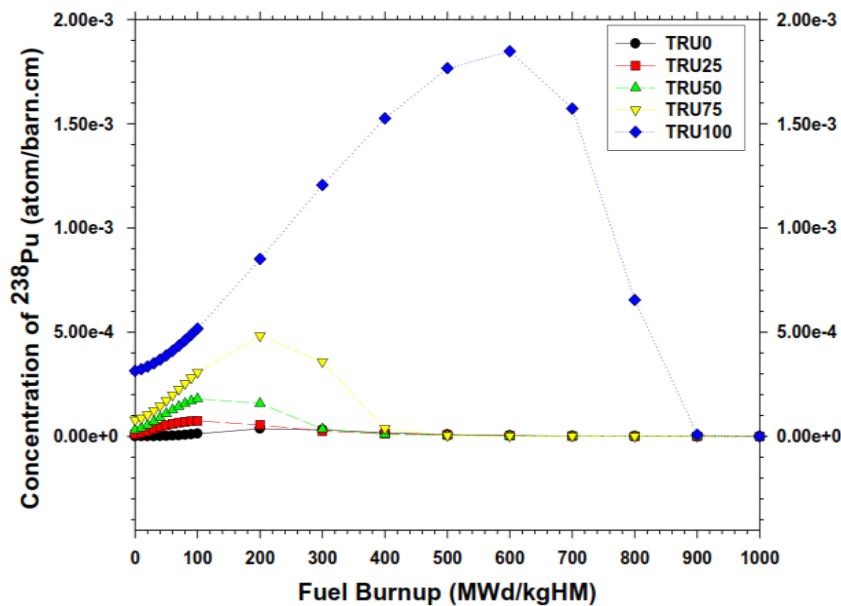


Fig. 5 – Concentration of ^{238}Pu as a function of fuel burnup.

From the proliferation standpoint, the discussion is more relevant to fissile plutonium of ^{239}Pu and ^{241}Pu which evolve during reactor operation. It is produced and burned within the reactor and considers that the reactor approaches a subcritical condition, namely within 70–500 MWd/kgHM, most of the fissile and fertile plutonium is at a significantly denatured composition. It prevents plutonium diversion

into nuclear weapons, even though the work of crushing the pebble and TRISO layers to extract plutonium is already very complex and prohibitively expensive.

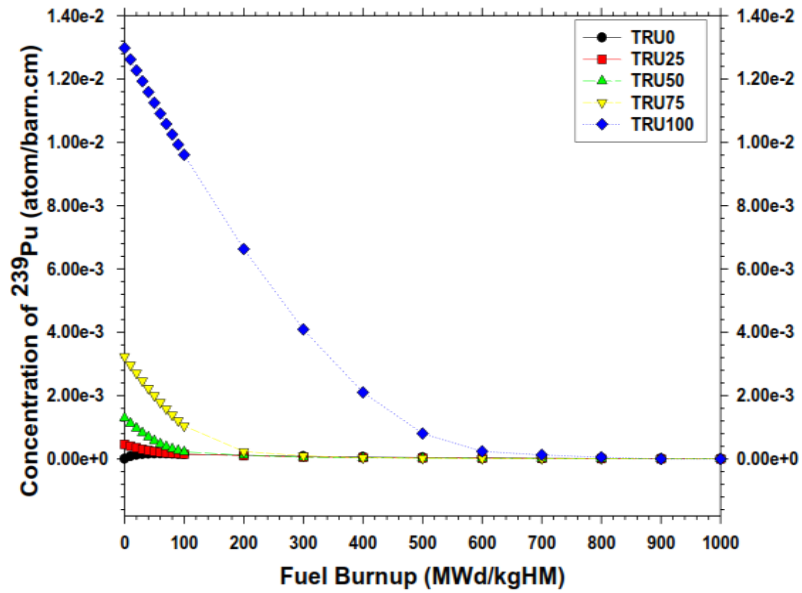


Fig. 6 – Concentration of ^{239}Pu as a function of fuel burnup.

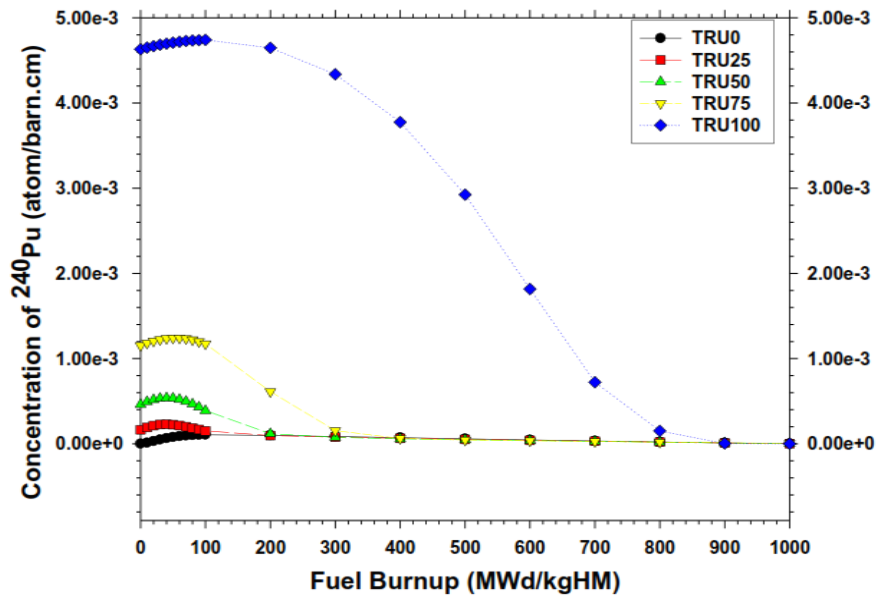


Fig. 7 – Concentration of ^{240}Pu as a function of fuel burnup.

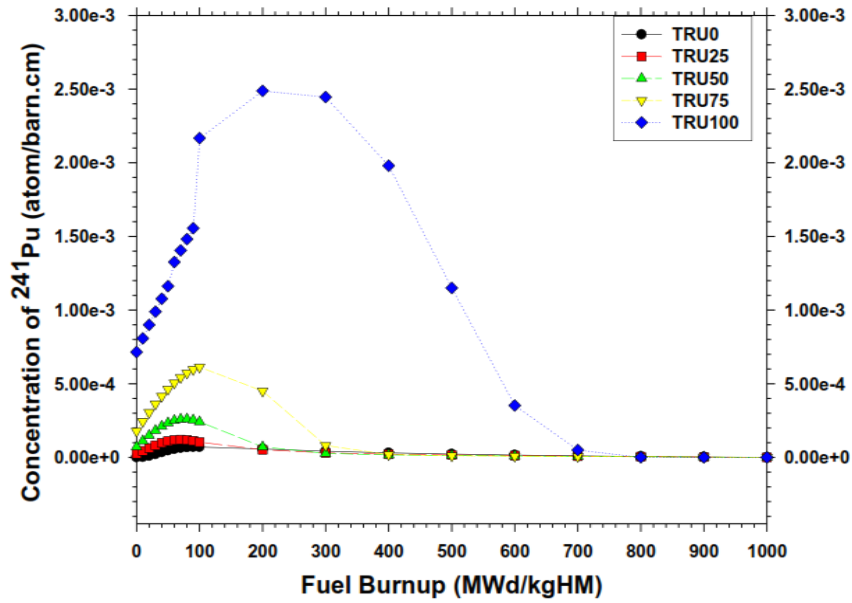


Fig. 8 – Concentration of ²⁴¹Pu as a function of fuel burnup.

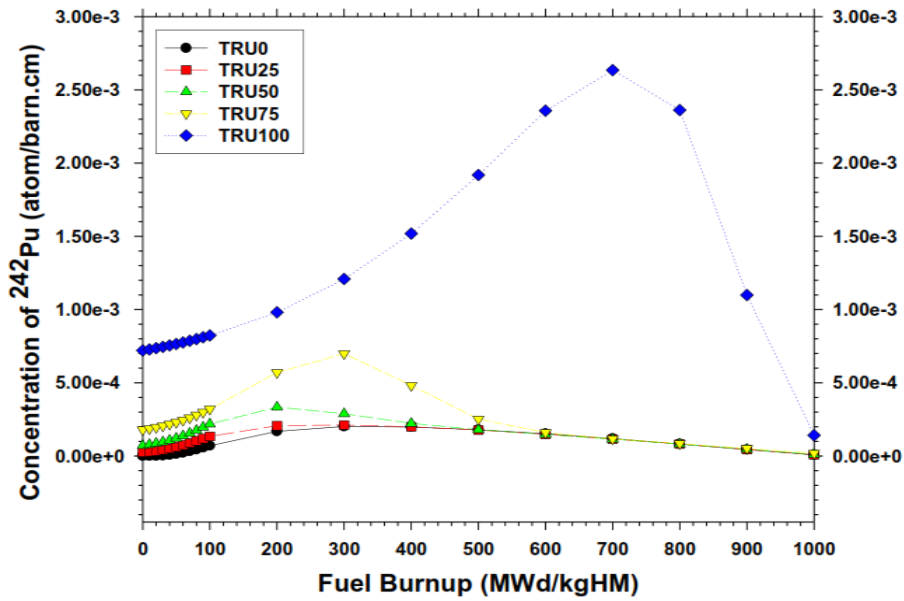


Fig. 9 – Concentration of ²⁴²Pu as a function of fuel burnup.

Minor Actinide Transmutation. Figures 10–13 illustrate the change in minor actinide isotopes concentration as a function of fuel burnup, ²³⁷Np, ²⁴¹Am, ²⁴³Am,

and ^{244}Cm respectively. In Fig. 10, it is shown that the ^{237}Np concentration at the beginning of cycle is higher in a core with a full TRU fuel pebble (TRU100 core) rather than a UO_2 fuel pebble.

The ^{237}Np will deplete in longer time in higher TRU content cores since higher TRU means higher ^{237}Np content. In UO_2 fuel pebble, ^{237}Np is produced by non-fissionable capture of ^{235}U and its subsequent capture reactions. It can also be resulted from the (n,2n) reaction of ^{238}U which then undergoes β decay. Because the reactor power is constant, the ^{237}Np concentration will reach an equilibrium state between the production rate and consumption rate (decay and capture), and finally, it will decrease since ^{238}U will degrade which will directly decrease the production rate.

For other isotopes such as Am and Cm, their evolution mechanisms are not different from plutonium since both are predominantly produced by neutron capture and β decay of their lower atomic number isotopes. The trend is found for the ^{241}Am concentration in Fig. 11, which is more or less similar to ^{239}Pu in Fig. 6 because it captures two neutrons and then undergoes β decay into ^{241}Am . The difference is that ^{241}Am concentration is lower than ^{239}Pu since only a portion of it experiences a neutron capture twice. The ^{243}Am concentration in Fig. 12 is like ^{242}Pu because it could capture a neutron and undergoes β decay to ^{243}Am , but because a fraction of it also underwent neutron capture, its concentration is smaller than ^{242}Pu . The peak concentration of this isotope is in the latter stage of burnup compared to their mother isotope.

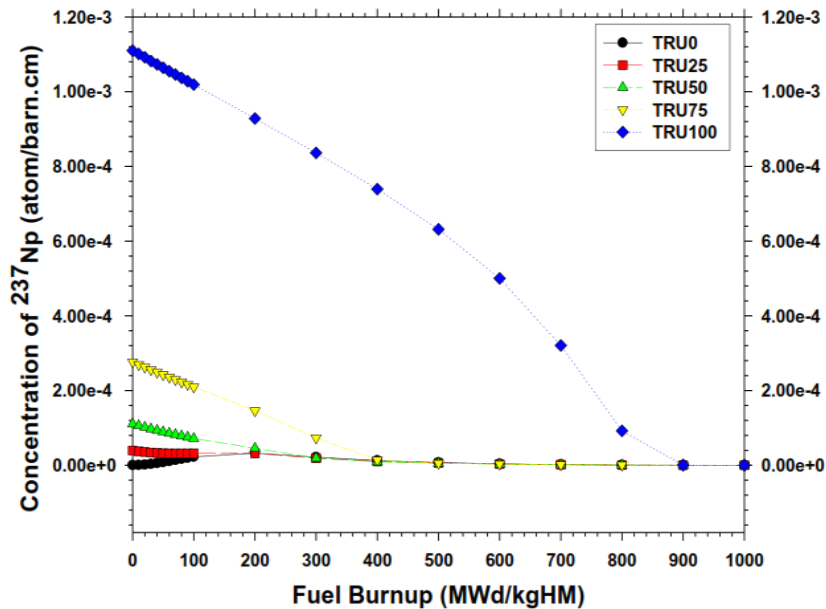


Fig. 10 – Concentration of ^{237}Np as a function of fuel burnup.

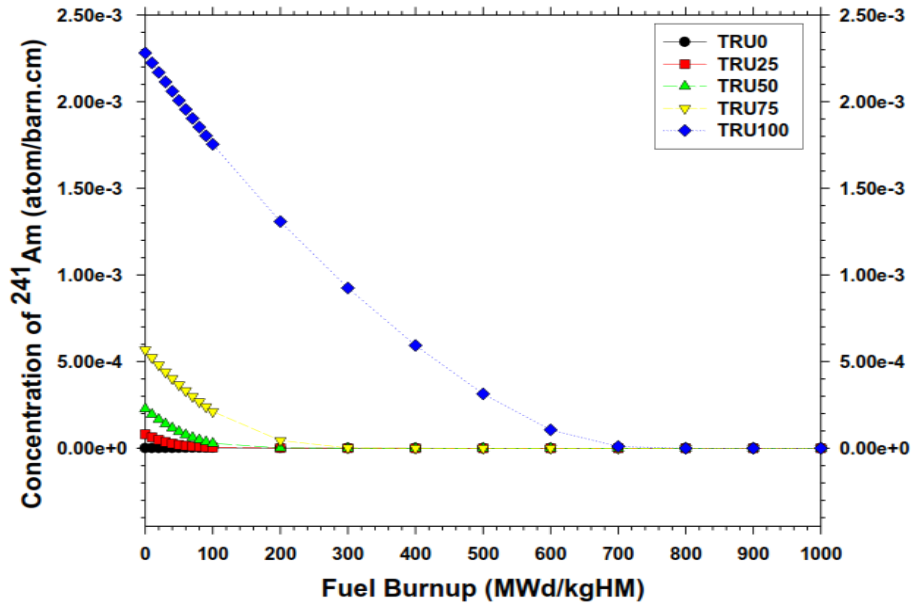


Fig. 11 – Concentration of ²⁴¹Am as a function of fuel burnup.

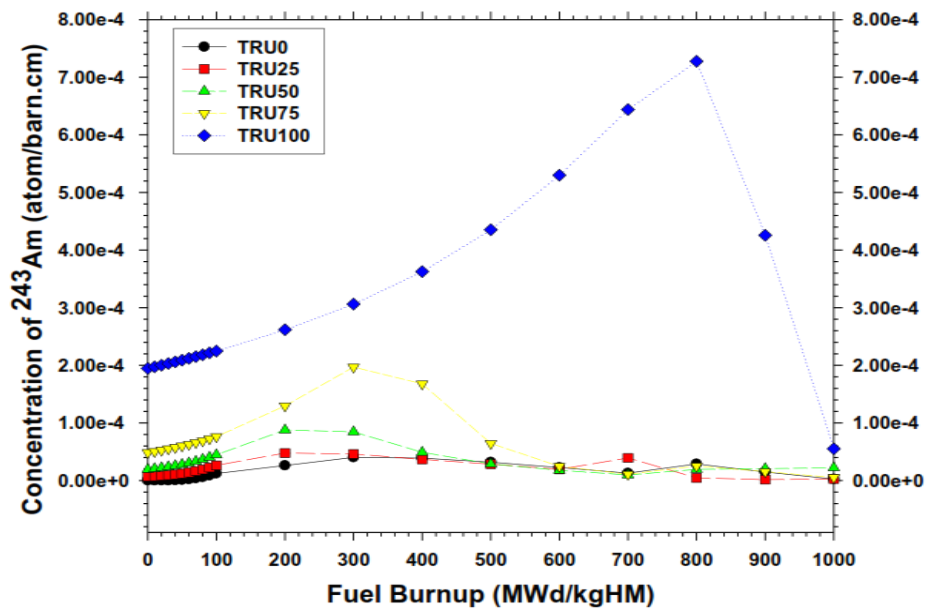


Fig. 12 – Concentration of ²⁴³Am as a function of fuel burnup.

The same thing happened for ²⁴⁴Cm concentration in Fig. 13 which resulted from neutron capture and β decay of ²⁴³Am. The ²⁴⁴Cm peak appears later than

^{243}Am with ^{244}Cm having a higher peak concentration. It is due to differences in the β decay rate of ^{244}Am to ^{244}Cm , ^{244}Cm formation rate, and the capture reaction of the ^{244}Cm isotope. The ^{244}Cm isotope formed from β -decay of ^{244}Am with 10.1 h half-life, while the α -decay of ^{244}Cm has 18.1 y of half-life.

Compared to ^{243}Am from β decay of ^{243}Am with a half-life of 4956 h, and ^{243}Am undergoing α decay with a half-life of 7370 y, the accumulation of ^{243}Am should be greater than ^{244}Cm . However, with ^{243}Am capture cross-section (n, γ) of 79.26 b and ^{244}Cm capture cross-section of 15.24 b (5 times lower) in the thermal neutron spectrum, the capture reaction rate of ^{243}Am will be greater than ^{244}Cm , unless if concentration of ^{244}Cm is 5 times greater than ^{243}Am .

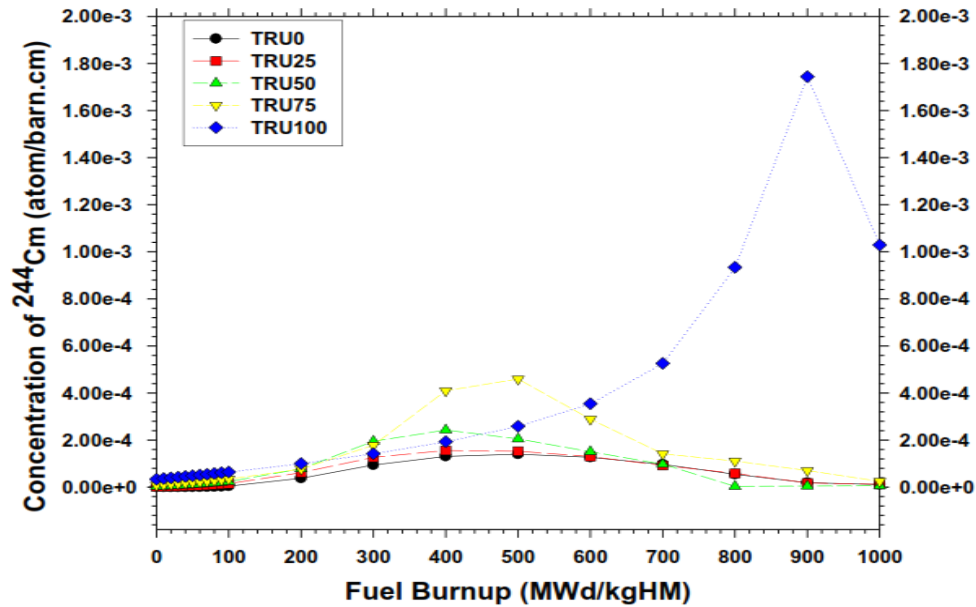


Fig. 13 – Concentration of ^{244}Cm as a function of fuel burnup.

The transmutation of TRU fuel in core configuration with full TRU fuel pebble (TRU100 core) is summarized in Table 8. The transmutation fraction was calculated as a mass reduction of the end of cycle (EOC) from the beginning of cycle (BOC). As shown above, the mass of the minor actinide isotope of ^{243}Am and ^{244}Cm increase with fuel burnup whereas ^{237}Np and ^{241}Am decrease with fuel burnup. Meanwhile, for plutonium isotopes, the mass of ^{238}Pu , ^{241}Pu , and ^{242}Pu are increased, while ^{239}Pu and ^{240}Pu are decreased. ^{241}Am is a long-lived radioisotope, aside from ^{239}Pu which is the main contributor to spent fuel radioactivity. In this study, they can be reduced significantly up to ~93.80% and ~86.25%. ^{240}Pu is the second largest contributor to radiotoxicity and together with ^{237}Np can be reduced through transmutation significantly up to a fraction of ~36.86% and ~43.10%, respectively.

However, the accumulation of ^{243}Am and ^{244}Cm slightly degrades these advantages despite their concentrations being relatively small. Similarly, ^{238}Pu , ^{241}Pu , and ^{242}Pu mass are increased for $\sim 461.8\%$, $\sim 60.91\%$, and $\sim 166.2\%$, respectively, which are not particularly offsetting the TRU mass reduction as their initial composition in the PWR spent fuel is quite small. ^{242}Pu is also the least radiotoxic isotope of plutonium. Hence, TRU mass reduction after 500 MWd/kgHM is 1.950×10^5 g. These results demonstrate that a pebble bed reactor with full PWR spent fuel can reduce TRU mass through transmutation up to 55.47% of its initial mass.

Table 8

Transmutation rate for TRU100 core

Isotope	Mass at 0 MWd/kgHM (g)	Mass at 500 MWd/kgHM (g)	Mass reduction after 500 MWd/kgHM (g)	Transmutation fraction (%)
^{237}Np	1.706×10^4	9.708×10^3	7.353×10^3	43.10
^{238}Pu	4.855×10^3	2.728×10^4	-2.242×10^4	-461.8
^{239}Pu	2.012×10^5	1.247×10^4	1.887×10^5	93.80
^{240}Pu	7.210×10^4	4.553×10^4	2.658×10^4	36.86
^{241}Pu	1.117×10^4	1.798×10^4	-6.805×10^3	-60.91
^{242}Pu	1.132×10^4	3.013×10^4	-1.881×10^4	-166.2
^{241}Am	3.565×10^4	4.903×10^3	3.075×10^4	86.25
^{243}Am	3.070×10^3	6.860×10^3	-3.790×10^3	-123.5
^{244}Cm	5.353×10^2	4.100×10^3	-3.565×10^3	-666.0
Total	3.569×10^5	1.589×10^5	1.980×10^5	55.47

5. CONCLUSION

Investigation on neutronic behavior of a pebble bed reactor for TRU transmutation has been performed using MCNP6 radiation transport code with ENDF/B-VII.1 nuclear data library. From the calculations, it can be observed that initial k_{eff} values decrease as the number of TRU fuel pebbles increases in the core. An increasing TRU fuel pebble ratio results in smaller DTC and larger MTC. The β_{eff} values decrease with the increased TRU fuel fraction, as well as other kinetic parameters. The calculated mass of plutonium and minor actinide demonstrate that a pebble bed reactor with 100% TRU from PWR spent fuel can reduce its TRU content for up to 55.47%. It can be concluded that the pebble bed reactor can be used to overcome the TRU accumulation produced from spent LWR fuel with several drawbacks on the neutronic behavior of reactivity coefficient and kinetic parameters, which could lead to changing the mechanism to control neutron population safely.

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