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ANALYSIS OF VARIATION MINOR ACTINIDE PIN CONFIGURATIONS Np-237, AM-241, AND Cm-244 IN UN-PuN FUELED PRESSURIZED WATER REACTOR

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Abstract

Actinide minor is a reactor waste with high toxicity and a long half-life. Minor actinides can be reduced by reusing them as fuel mixtures in reactors. This research uses PWR reactors with the primary fuel UN-PuN or Uranium Plutonium Nitride with a burning time of 5 years. The fuel consists of enriched Uranium, reactor-grade Plutonium from LWR waste, and minor actinides including Neptunium-237, Americium-241, and Curium-244. The purpose of this study was to find a design that is effective in reducing minor actinide waste. There are six designs or cases used in the addition of minor actinides. Each case has six minor actinide pins in each assembly. The addition of minor actinides is arranged in heterogeneous cores. The analysis was carried out by observing the values of k-eff, excess reactivity, and mass of minor actinides obtained from simulations using OpenMC code 0.13.2 and the ENDF/B-VIII library. The homogeneous core obtained an excess reactivity of 9.7 % with a percentage of plutonium of 8 %. The results of the homogeneous core are used as a reference for preparing a heterogeneous core. The heterogeneous core obtained an excess reactivity of 9.9 % with a percentage of plutonium $F1$: 5.5 %, $F2$: 8 %, and $F3$: 10.5 %. *Np*-237 can be reduced by 53 kg, and *Am*-241 can be reduced by 61 kg with minor actinide pins in case 1. *Cm*-244 can be reduced by 363 kilograms with minor actinide pins in case 6. Excess reactivity in the addition of *Np*-237 and *Am*-241 decreased to 5.3 %, while the accumulation of *Cm*-244 increased to 12.1 %.

Keywords: excess reactivity, k-eff, mass, PWR, UN-PuN, OpenMC, pins minor actinide.

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1. Introduction

Minor actinides are reactor wastes with high levels of toxicity and long half-lives of up to 2,140,000 years. Minor actinides generated from reactor waste can include Neptunium (Np), Americium (Am), and Curium (Cm) [1]. Managing this waste is crucial for reactor safety. Minor actinides can be recycled by mixing them with primary reactor fuel. Through fuel burn-up in the reactor, minor actinides can be transformed into less toxic materials with shorter half-lives [2, 3].

PWR is a thermal-type reactor that uses cooling in the form of water and has a moderator to slow down the rate of neutrons. *PWR* has advantages in terms of stability, where this reactor tends to produce less power when there is an increase in temperature [4]. *PWR* there are three cooling systems used in the PWR reactor, namely the primary cooling system which is located on the reactor core, the secondary cooling system which is located on a pipe outside the reactor core, and the tertiary cooling system which is located on the condenser [5].

Fission reactions occurring in a PWR reactor involve thermal neutrons. Fission reaction is the process of splitting atomic nuclei into smaller nuclei when they collide with neutrons. This process yields lighter atomic nuclei, 2 to 3 neutron particles, and energy of approximately 200 MeV in gamma rays, which generates heat in the surrounding [6]. Not all neutrons interacting with the fuel undergo fission; only fissile materials such as U-235, Pu-239, and Pu-240 can undergo fission reactions [7]. Neutrons that are not utilized for fission may leak out of the reactor core or be absorbed by fertile materials. The comparison between the number of fission-produced neutrons and neutrons lost due to leakage or absorption indicates the critical parameter of the reactor known as the effective multiplication factor (k-eff) [8]. The value of k-eff consists of three conditions representing the reactor criticality: subcritical, critical, and supercritical.

Previous researchers have reviewed PWR research related to reactor waste analysis using simulation programs [9–12]. This research uses the OpenMC program to perform simulation calculations. OpenMC uses the Monte Carlo method that simulates neutron transport as a stochastic process, where the movement of the resulting neutrons will occur randomly [13]. OpenMC has the advantage of being open source to be used publicly. OpenMC was developed in 2011 by Computational Reactor Physics at the Massachusetts Institute of Technology [14]. OpenMC has a calculation system that uses a combination of filters and values. Each filter limits calculations based on attribute particles [15]. Filters can limit scoring to particles moving within a certain cell or pre-collision energy range [16].

This research carried out homogeneous and heterogeneous core calculations on *PWR* reactors using the OpenMC program. The main fuel for the PWR reactor consists of uranium plutonium nitride which has a melting point of 2500 °C [17]. *UN-PuN* fuel is mixed with minor actinide materials for various configurations. The goal is to find the configuration of minor actinide pins most effective for reducing the number of minor actinides worldwide. In addition, to determine the comparison of the acquisition of minor actinide pin configurations Neptunium (Np-237), Americium (Am-241), and Curium (Cm-244) in the Pressurized Water Reactor (*PWR*).

2. Materials and methods

The research procedure was carried out based on the flow diagram in **Fig. 1**. The research began by determining the research topic and conducting a literature review of nuclear reactors to identify problems that could be solved. The reactor specifications used are in accordance with the parameters in **Table 1**. The simulation calculations for this research use the neutron transport program in the form of OpenMC. Calculations of k-eff and mass of minor actinides in OpenMC were carried out using 300 batches, 30 inactives, and 50,000 particles. First of all, carry out calculations for the reactor core with the same (homogeneous) fuel composition with a number of variations in 11 cases. Data with the optimal criticality value (1 case) was taken for further calculations, namely on the heterogeneous reactor core (3 types of fuel composition) with a total variation of 6 cases. Next, the optimal data (1 case) was taken to be used as a reference in calculating the addition of minor actinides in pin form. The minor actinide pin design used has 6 cases in which each pin point is arranged differently (**Fig. 3**). The addition of minor actinides will result in a stable decrease in the criticality value at each point in the combustion time and approaching critical conditions (k-eff≈ 1). Apart from that, it can also minimize the mass of minor actinide waste (Np-237, Am-241, Cm-244) which has toxic characteristics and a long half-life. Finally, a minor actinide pin design will be obtained that is able to reduce the most mass Np-237, Am-241, and Cm-244 waste in PWR reactors.

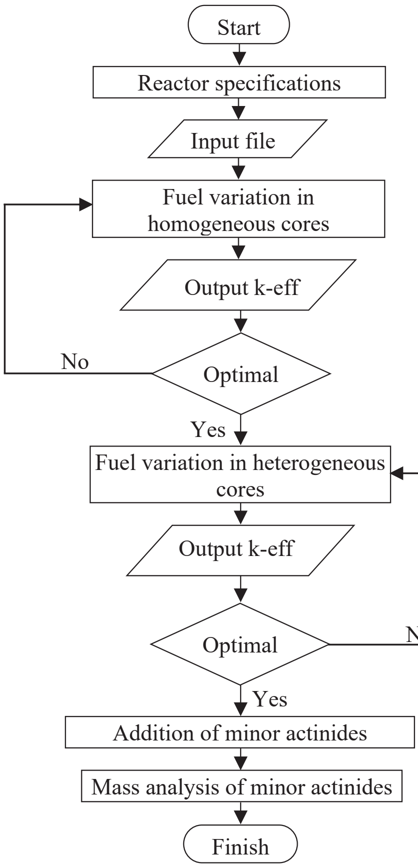


Fig. 1. Research flow chart

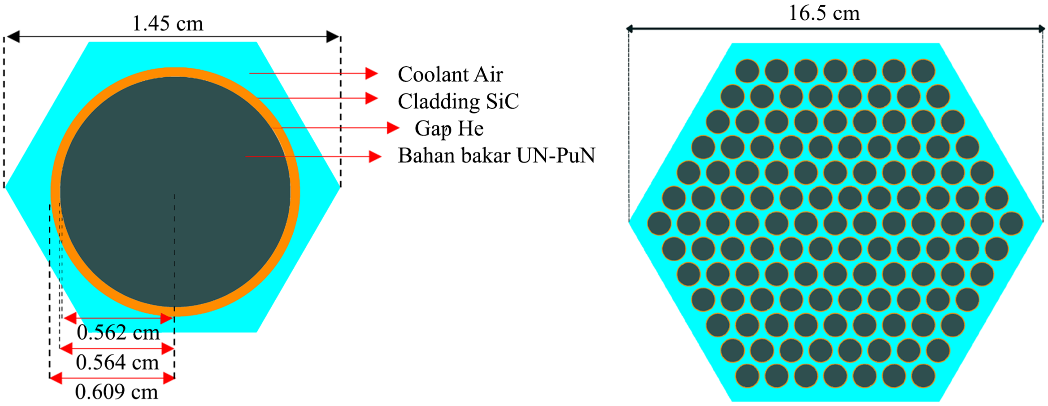
The reactor design specifications used in this study are listed in **Table 1**. The reactor design consists of several parameters generally found in *PWR* reactors. These parameters are reactor power, burn-up period, shape and size of the reactor core, and fuel used. This research used uranium plutonium nitride (UN-PuN) fuel. The uranium used consists of 0.7 % *U*-235 and 99.3 % *U*-238. The plutonium used is waste from the Light Water Reactor (LWR), which has burn-up 33 GWd/ton. The plutonium material consists of several isotopes, including Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242. The percentage of plutonium isotopes respectively is 1.8 %, 58.7 %, 24.2 %, 11.4 %, and 3.9 % [18].

Table 1

Reactor design specifications

Parameter	Specifications
Power (MWt)	300
burn-up (day)	1825
Core Geometry	Cylindrical Pancakes
Pin type	Hexagonal
Core height (cm)	100
Core diameter (cm)	300
Reflector width (cm)	60
Absorber width (cm)	20
Fuel	Uranium Plutonium Nitrida (UN-PuN)
Cladding	*Silicon Carbida* (SiC)
Gap	Helium (He)
Reflector	Stainless Steel
Absorber	*Boron Carbide* (B_4C)

The reactor core is coated by the reflector and absorber materials axially and radially. Reflector and absorber materials have a density of 3.210 gr/cc and 2.52 gr/cc [19]. Uranium Plutonium Nitride (UN-PuN) fuel is arranged in hexagonal fuel pins. The fuel pin consists of a UN-PuN fuel layer, Helium gap (He), Silicon Carbide cladding (SiC), and water cooling (H_2O). The volume fraction of each fuel pin consists of 60 % fuel, 0.5 % gap, 10 % cladding, and 25 % coolant. Fuel pins are arranged in assemblies, each consisting of 127 fuel pins. The arrangement of fuel and assembly pins can be seen in **Fig. 2**.



a *b*

Fig. 2. Fuel reactor arrangement: *a* – Fuel pins arrangement; *b* – Assembly pins arrangement

The minor actinide waste used is arranged in 6 designs, as shown in **Fig. 3**. The minor actinides used are Neptunium (Np-237), Americium (Am-241), and Curium (Cm-244). Minor actinides are formed within separate fuel pins as shown in **Fig. 2** and distributed according to **Fig. 3**. Each minor actinide is burned up separately for each case to observe its individual characteristics.

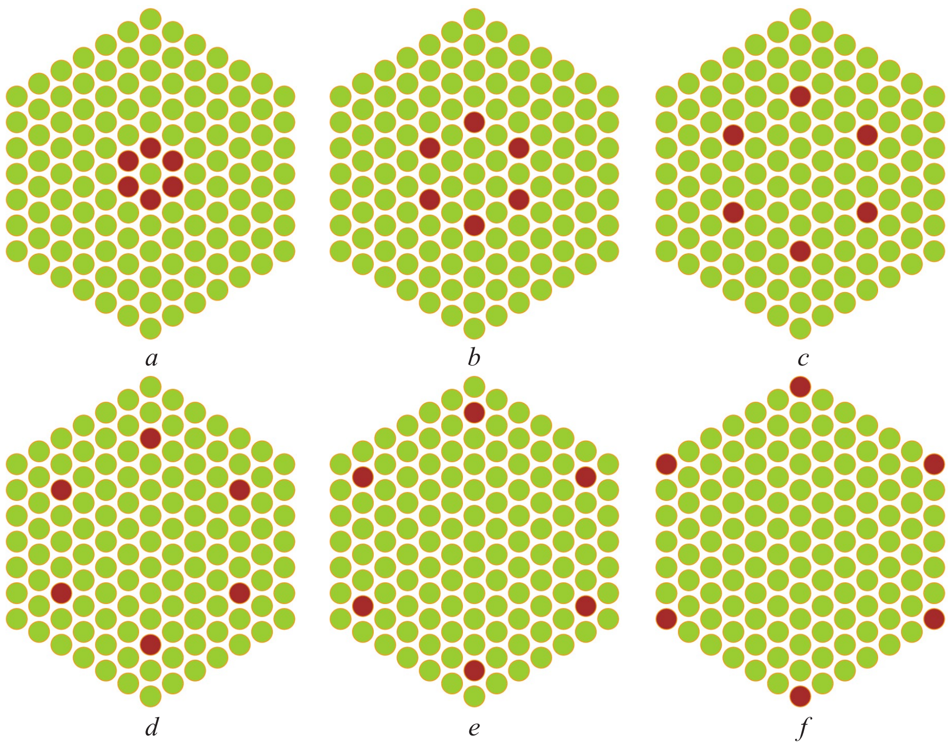


Fig. 3. Variation Minor Actinide Pin Configuration: *a* – Case 1; *b* – Case 2; *c* – Case 3; *d* – Case 4; *e* – Case 5; *f* – Case 6

The analysis was carried out by observing the values of k-eff, excess reactivity, and mass of minor actinides obtained from simulations using OpenMC code 0.13.2 and the ENDF/B-VIII library. The fuel must be changed to mole fractions before being used on OpenMC. Changing the fuel is done using formula (1). Data obtained from OpenMC are entered in the Origin software to obtain excess reactivity values, small actinide mass, and graphing. The value of excess reactivity is obtained using formula (2). Analysis of k-eff values and excess reactivity is performed to determine the optimal design on homogeneous and heterogeneous nuclei. Mass analysis of minor actinides is performed by unit conversion from particle to mass using formula (3):

$$\text{Mass fraction } X = \frac{Mr\,X \times \text{Mol fraction } X\ (\%)}{\text{Total mass}} \times 100\,\%, \quad (1)$$

$$\text{Excess Reactivity} = \frac{k\text{-eff} - 1}{k\text{-eff}} \times 100\,\%, \quad (2)$$

$$\text{Mass (kg)} = \frac{n}{6.02 \times 10^{23}} \times \frac{Mr}{100}, \quad (3)$$

where Mr – relative molecular mass; k-eff – effective multiplication factor value; n – number of atoms.

3. Results and discussion

This research used uranium plutonium nitride (UN-PuN) fuel. The uranium used consists of 0.7 % U-235 and 99.3 % *U*-238. The plutonium used is waste from the Light Water Reactor (LWR), which has burn-up 33 GWd/ton. The plutonium material consists of several isotopes, including Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242. The percentage of plutonium isotopes respectively is 1.8 %, 58.7 %, 24.2 %, 11.4 %, and 3.9 % [19]. The volume fraction of each fuel pin consists of 60 % fuel, 0.5 % gap, 10 % cladding, and 25 % coolant. The calculation of k-eff and mass of minor actinides in OpenMC was done using 300 batches, 30 inactives, and 50,000 particles. The calculation in this study was only done for 1825 days or 5 years. The minor actinides used in this study were Np-237, Am-241, and Cm-244. This study aims to obtain the most effective minor actinide pin design to reduce the amount of minor actinides in PWR reactors.

The configuration of a homogeneous core is carried out with plutonium variations of 5 % to 15 %. **Fig. 4** shows k-eff values on homogeneous cores over a 5-year burn-up period.

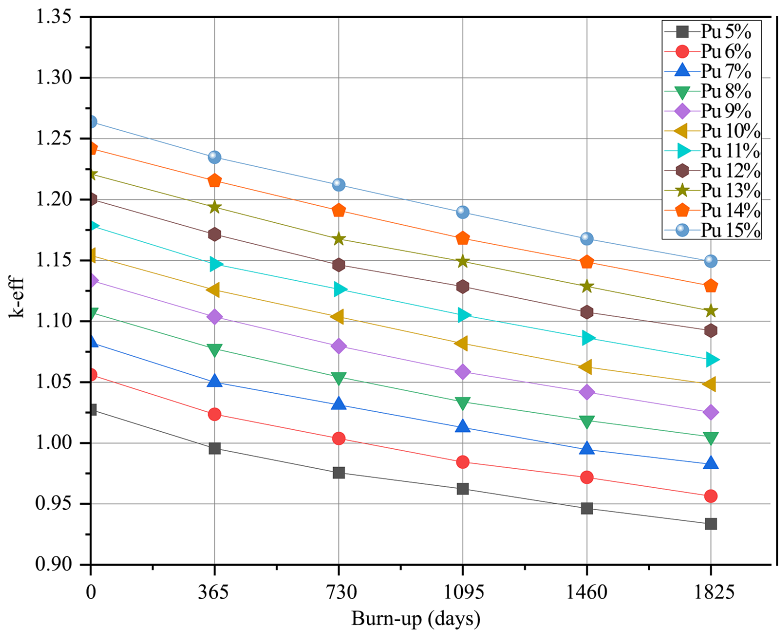


Fig. 4. The value of k-eff on homogeneous core

The graphic trend of the k-eff value is obtained from stochastic simulation calculations by OpenMC using formula (2). The k-eff value is calculated once every year in a cycle of 5 years or 1825 days. Plutonium with a percentage of 8 % is close to critical k-eff, and this variation is used to prepare heterogeneous core. The maximum excess reactivity obtained is 9.7 %.

Heterogeneous core is arranged with three different percentages of plutonium. **Fig. 5** is an arrangement of heterogeneous core. The highest percentage of plutonium is placed at $F3$, and the lowest is placed at $F1$. The percentage variation of plutonium used in heterogeneous cores is shown in **Table 2**.

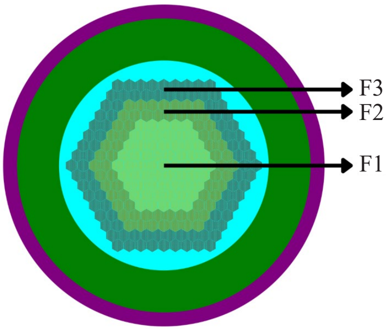


Fig. 5. Fuel arrangement on heterogeneous cores

Table 2
The percentage of plutonium in heterogeneous cores

No.	Plutonium percentage			Average
	***F*1**	***F*2**	***F*3**	
1	5 %	8 %	11 %	8 %
2	5.5 %	8 %	10.5 %	8 %
3	6 %	8 %	10 %	8 %
4	6.5 %	8 %	9.5 %	8 %
5	7 %	8 %	9 %	8 %
6	7.5 %	8 %	8.5 %	8 %

Fig. 6 shows the k-eff obtained in each case during the 5-year burn-up period. The k-eff with $F1$:5.5 %, F2:8 %, and F3:10.5 % has a value that is close to critical. The maximum excess reactivity obtained is 9.9 %. This case was used to add minor actinides with plutonium percentages $F1$:5.5 %, $F2$:8 %, and $F3$:10.5 % on heterogeneous cores.

Minor actinides were added to 6 different assembly cases, as shown in **Fig. 3**. **Fig. 7** shows the k-eff obtained after the addition of minor actinides. The optimal addition of Np-237 and Am-241 occurs in case 1. The approximate critical condition can be maintained for up to 730 days and enters sub-critical conditions in the next burn-up until the last year (1825 days). The maximum excess reactivity when adding Np-237 and Am-241 has the same value of 5.3 %. Meanwhile, for the addition of Cm-244, the optimal situation is in case 6, where the k-eff can approach the critical condition for 1825 days with a maximum excess reactivity of 11.8 %.

The k-eff obtained with each addition of minor actinides as shown in **Fig. 7** exhibits different characteristics. One of the most influential factors contributing to these differences is the value of the macroscopic cross section, which indicates the ability of each nuclide to absorb neutrons. **Fig. 8** illustrates the absorption cross sections for Np-237, Am-241, and Cm-244. Np-237 and Am-241 have higher neutron absorption probabilities at low energies compared to Cm-244. This reduces the neutron population available for fission, resulting in a significant decrease in k-eff due to the addition of Np-237 and Am-241 [20].

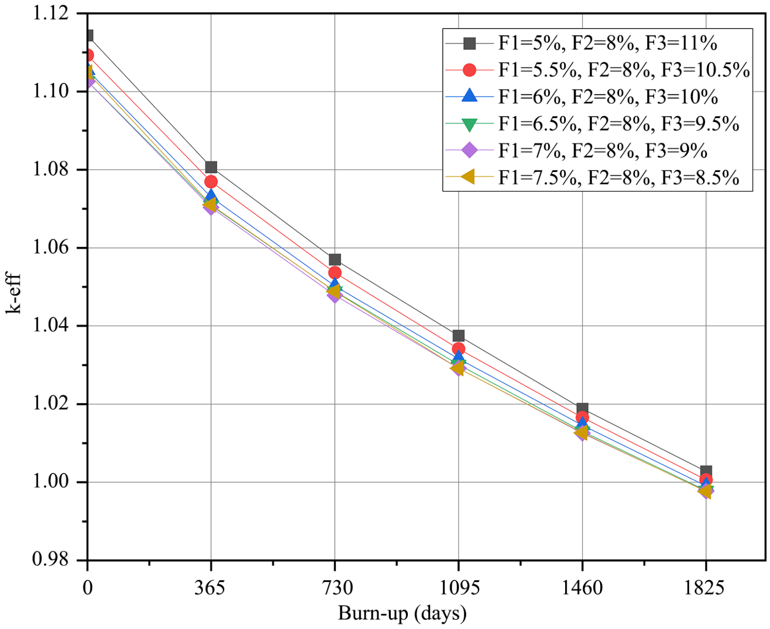


Fig. 6. The value of *k*-eff on heterogeneous cores

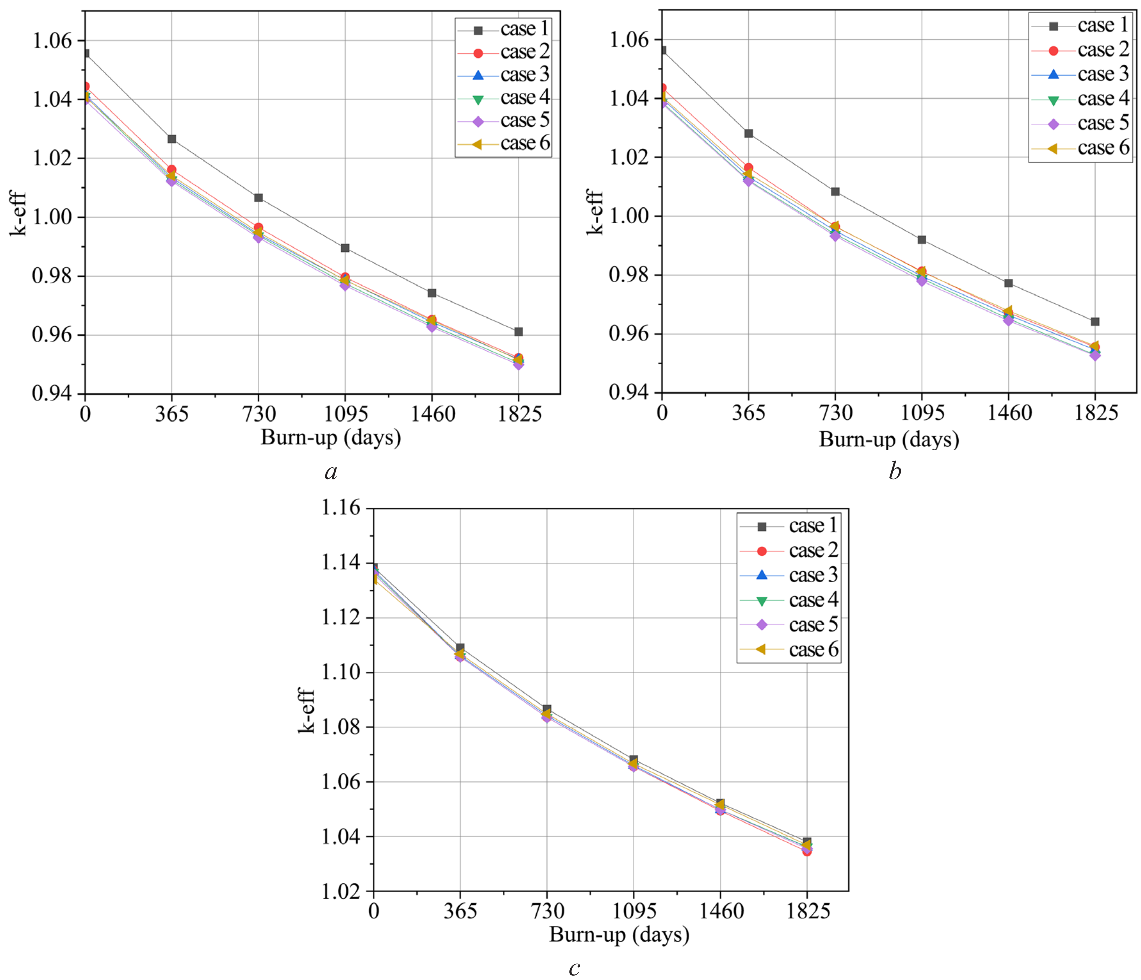


Fig. 7. Characteristic value of *k*-eff: *a* – the *k*-eff value in addition to *Np*-237; *b* – the *k*-eff value on the addition of *Am*-241; *c* – the *k*-eff value in addition to *Cm*-244

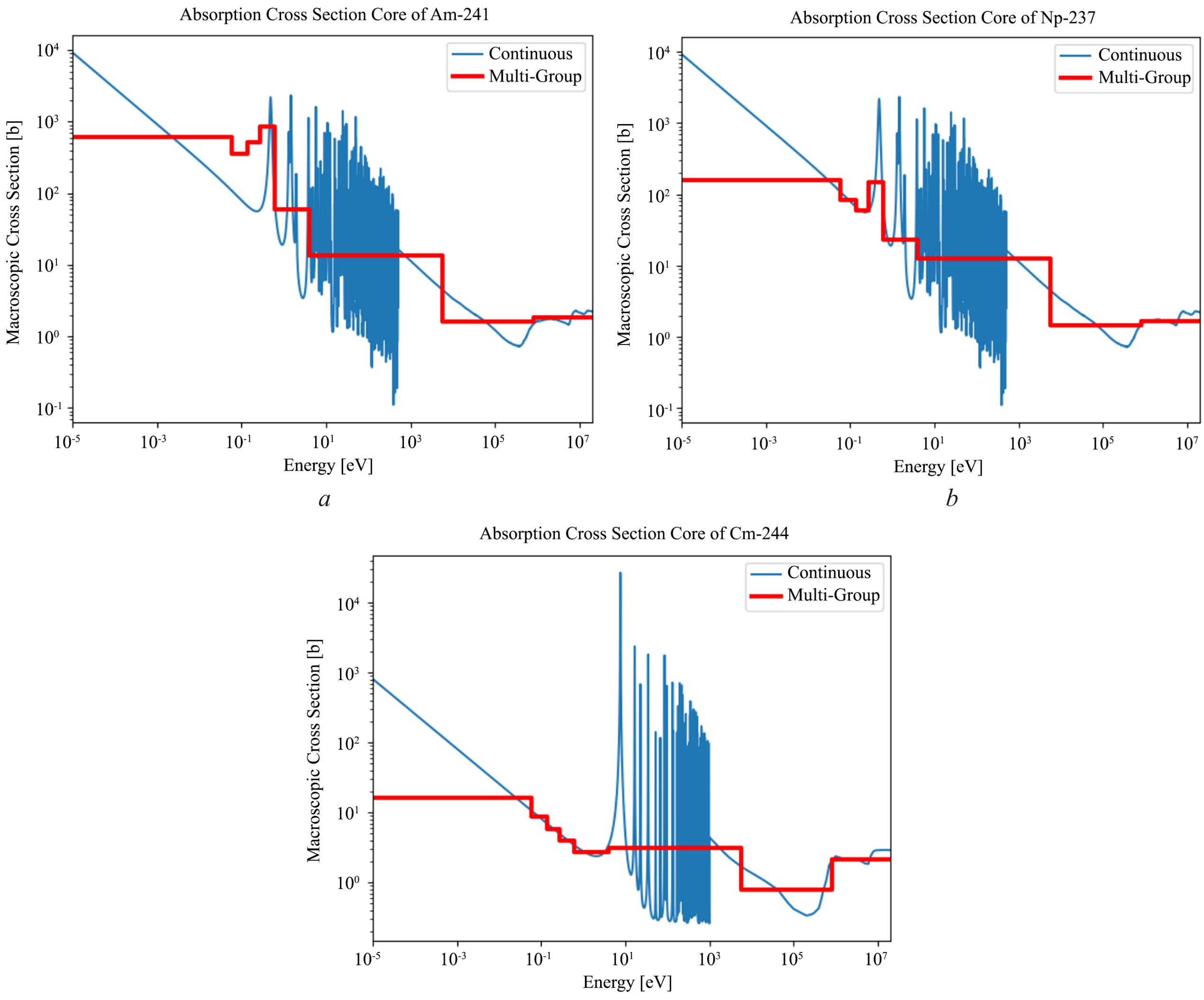


Fig. 8. Characteristics of the macroscopic cross section absorption: *a* – absorption cross section Np-237; *b* – absorption cross section Am-241; *c* – absorption cross section Cm-244

Fig. 9 shows the mass of minor actinides (Np-237, Am-241, and Cm-244) after being added to the reactor core.

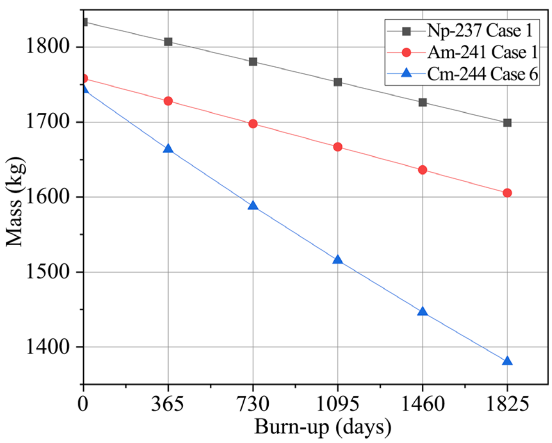


Fig. 9. Mass comparison of Np-237, Am-241, and Cm-244

The mass of each minor actinide decreases every day. Np-237 was reduced by 53 kg from 1833.74 kg to 1780.70 kg in 730 days. Am-241 was reduced by 61 kg from 1758.39 kg to 1697.70 kg also in 730 days. Cm-244 was reduced by 363 kg from 1743.37 kg to 1380.28 kg in 5 years burn-up. Mass calculations of Np-237 and Am-241 were taken when they were in a critical state for up to 730 days. Nuclides that absorb neutrons will allow them to become other nuclides.

Minor actinide materials Np-237, Am-241, Cm-244 have the opportunity to decay into fertile materials in the form of Pu-238 and Pu-240. Np-237 and Am-241 have the possibility of being Pu-238, while Cm-244 has the potential of being Pu-240, as shown in **Fig. 11** which is the cycle of isotopic decay of nuclear material. Pu-238 and Pu-240 are fertile materials. Fertile material helps produce fissile material in reactors. The decay results will cause a change in the mass of the minor actinide material produced, as shown in **Fig. 10.**

Graph in **Fig. 10** represents the mass of plutonium over a 5-year burn-up period. The mass of plutonium is increasing every day. The Pu-240 derived from Cm-244 significantly improved over Np-237 and Am-241. It is in harmony with the mass of Cm-244, which is also a lot of reduction.

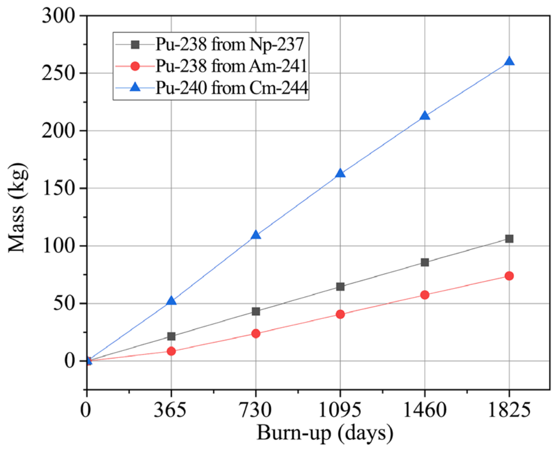


Fig. 10. Mass comparison of plutonium from the addition of minor actinides

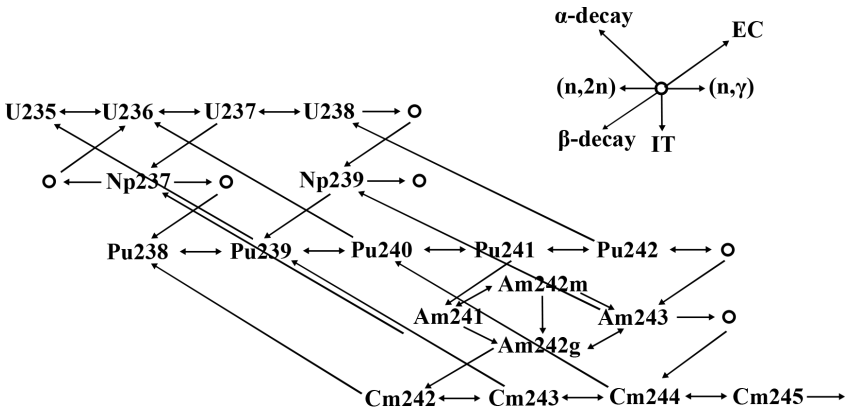


Fig. 11. Nuclide decay chains [21]

4. Conclusions

The conclusion was that adding minor actinides in the form of pins can reduce the number of minor actinides Np-237, Am-241, and Cm-244. Np-237 can be reduced by 53 kg using the case 1 design, producing a maximum excess reactivity of 5.3 %. Am-241 can be reduced by 61 kilograms using case 1 design, with a maximum excess reactivity of 5.3 %. Cm-244 can be reduced by 363 kg using the case 6 design, producing a maximum excess reactivity of 11.8 %.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

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Data availability

Data will be made available on reasonable request.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

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