

The Effect of Adding Minor Actinide Fuel Rods on GFR Reactor in Radiopharmaceutical Waste Production Using OpenMC Program

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Abstract

GFR is a generation IV reactor based on helium gas refrigeration capable of working at very high temperatures. The fast spectrum in this reactor makes it possible to use nitride-based fuel, namely Uranium Plutonium Nitride (UN-PuN). Adding minor actinide (MA) material to the primary fuel, UN-PuN can maximize reactor performance to near critical from the beginning to the end of burn-up. This study aims to analyze the effect of adding MA fuel rods to the heterogeneous core of 5 fuel variations (F1, F2, F3, F4, F5) on the probability of radiopharmaceutical waste production. The method in this research is to place MA fuel rods in this study using four designs based on the highest neutron flux value in one fuel assembly. The results of the neutron flux calculation show that the reactor's active core's central region (F1, F2, F3) needs to be added to MA fuel rods so that the resulting flux is more evenly distributed. The calculation of reactor criticality shows that Np fuel rod design 4 and Am fuel rod design 1 have the best k_{eff} value ($k_{\text{eff}} \approx 1$) among other designs. The burn-up of MA fuel rods produces a minimal probability of producing Tc99m, Sr89, Y90, Rh105, Ag111, I231, and Sm15 radiopharmaceutical waste, even less than 1 kg.

Keywords

GFR, Neutron Flux, Radiopharmaceuticals, Waste Production

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1. INTRODUCTION

One type of reactor that uses fast neutrons in carrying out fission chain reactions and a combination of resources efficiency is GFR or Gas-cooled Fast Reactor (van Rooijen and Kloosterman, 2009). GFR is a generation IV revolution design planned to be realized in 2030 (Generation IV International Forum (GIF), 2002). Generation IV reactors are designed based on four main goals: sustainability, affordable costs, high level of security, and non-proliferation (Pioro, 2016). The main target of generation IV reactors is the transmutation of Minor Actinides (MAs) (Castelliti et al., 2009). GFR works at a very high-temperature system (helium temperature of 850°C), which allows it to produce hydrogen gas (Zohuri, 2020). Helium gas has the advantage of being single-phase and inert (Miletic et al., 2014). The reactor design reference is based on a GFR core with a capacity of 2400 MWth located in a high-pressure steel vessel (Lima-Reinaldo et al., 2019). GFR reactors use the same fuel recycling process as SFR and the same reactor technology system as VHTR (Billot and Seran, 2007). Fast neutron spectra in these reactors allow for receiving various types of

fuels, including Pu-based degradation from LWR spent fuel, MOX fuel, nitride, and Actinide Minor (Foley and Knight, 2009). This flexibility in fuel options is crucial for the efficient management of spent fuel and the development of sustainable nuclear energy systems, a necessity highlighted by the role of nuclear energy in contributing to achieving net-zero emissions (Permana et al., 2022).

More than 98.5% of waste fuels generally consist of short-lived uranium and fission products (Hu et al., 2015). About 1% of spent fuel comprises plutonium material and minor actinide isotopes (Madic et al., 2007). Actinide minor is a spent fuel from LWR reactor operation with high toxicity characteristics and a long decay rate of up to 2 million years (Syarifah et al., 2020). There are three types of minor actinides commonly produced by reactor cores, namely Neptunium (Np), Americium (Am), and Curium (Cm). The minor actinide nuclides with the most significant percentage produced from LWR reactor operating waste are Np237 (49.14%), Am241 (29.98%), Am243 (15.5%), and Cm244 (4.99%) (International Atomic Energy Agency (IAEA), 2009). The presence of minor actinides must be minimized because of their toxic environmen-

tal characteristics. Therefore, it is necessary to recycle minor actinides that can be used as the primary mixed fuel in reactors. The recycling process aims to convert minor actinide nuclides into nuclides that are more environmentally friendly or have faster decay characteristics. This process is commonly called the transmutation method (Takeda, 2016). The transmutation process resulting from the burn-up of minor actinides can have the opportunity to produce short-lived radioactive isotope waste, called radioisotopes, which is helpful for the needs of the health or radiopharmaceutical sector (Harries, 1974). Some radiopharmaceutical nuclide products include Tc99m, Sr89, Y90, Rh105, Ag111, I231, Sm153, and others (Holland et al., 2010).

Several researchers have used the transmutation or addition of minor actinides to the primary fuel. Ibrahim et al. (2017) in their research compared the transmutation parameters of Minor Actinide (MA) for the homogeneous core design of IC (Inner Core) and OC (Outer Core) in the GFR-2400 reactor fuel assembly. A total of 516 fuel assemblies presented active fuel regions consisting of two core zones, namely 264 fuel assemblies for IC and 252 for OC. The composition of Pu in IC is 14.12%, and OC is 17.65%. The calculation results show that the MA breeding rate in the OC fuel assembly is higher than that of the fuel assembly IC. The mass increase of MA in IC cores is 0.4%, and OC cores are 0.5%. However, this study did not treat the addition of MA fuel; it only analyzed the MA transmutation resulting from the Pu decay process. The addition of MA to the primary fuel in a GFR reactor involves the use of fast neutrons to induce fission reactions in the fuel, which can lead to the transmutation of MA into shorter-lived isotopes. This approach is essential for efficiently managing nuclear waste and developing sustainable nuclear energy systems (Syarifah et al., 2020). Takeda et al. (2017) examine the uncertainty of MA transmutation in fast reactors. The uncertainty of MA transmutation is calculated by evaluating the burn-up sensitivity to the JENDL-4 cross-section consisting of five terms: flux term, direct term, adjoint-flux term, power term, and number density term. The MA transmutation uncertainty method was applied to two fast reactor core designs with 6% and 11% MA content, respectively. The calculation results show uncertainty in the total MA transmutations of 3% and 4% for both core designs. This is due to the uncertainty of the transmutation of Np237 and Am241, which mainly contributes to the total transmutation, i.e., its value is minimal (about 3%). However, research for Cm244 material has considerable uncertainty (10% and 13%), so it needs to be considered when using MA fuel in the form of Cm244 from the recycling process. Significant uncertainties can produce large decay heat and neutron emissions.

Syarifah et al. (2020) in their research mixed minor actinide nuclides in the form of Americium (Am241 and Am243) and Neptunium-237 with UN-PuN fuel. The composition of each minor actinide material is varied by 0%-5%. The calculation results show that adding minor actinides can reduce the value of k_{eff} at the beginning of burn-up. However, the criticality value produced at the end of burn-up still tends to

be high, so it is necessary to add burnable poison material in the form of protactinium-231.

Prasetya et al. (2024) compared the fission product MA burn-up results of the GFR reactor fueled by UN-PuN on two heterogeneous core designs, namely three fuel variations and five fuel variations. Both designs have differences in the composition of Pu in each region of active fuel; the best composition of Pu in terms of criticality is for three variations in the form of F1 = 8%, F2 = 10%, and F3 = 12%. As for 5 fuel variations in the form of F1 = 8%, F2 = 9%, F3 = 10%, F4 = 11%, F5 = 12%. The calculation results show that the design of 5 fuel variations produces less MA product fission than three fuel variations. However, in research, the characteristics of criticality still tend to be high at the beginning of burn-up (BOL), so it is necessary to add an MA fuel rod.

Based on research that has been done (Ibrahim et al., 2017; Takeda et al., 2017; Syarifah et al., 2020; Prasetya et al., 2024), the characteristics of MA transmutation need to be further analyzed, especially in Gas Cooled Fast Reactor type. Therefore, the primary objective of this study is to investigate the effect of the addition of MA fuel rods from the recycling process on the generation of radiopharmaceutical burn-up waste. Using recycled MA fuel is expected to shorten the T1/2 period (decay rate) to make it more environmentally friendly.

2. EXPERIMENTAL SECTION

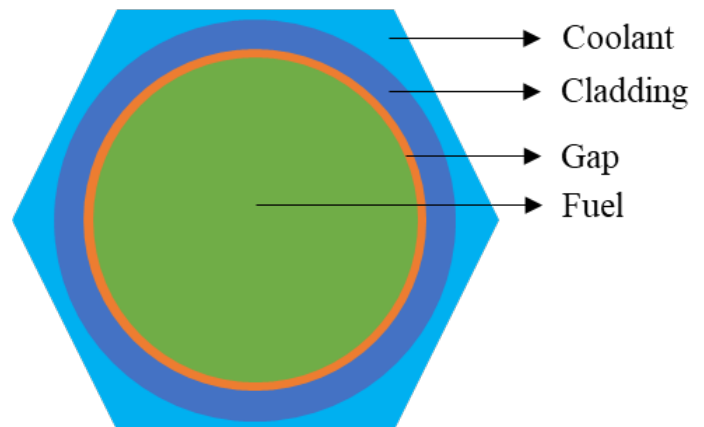


Figure 1. Fuel Pins Configuration

2.1 Materials

This research is a more comprehensive follow-up analysis than previously conducted by Prasetya et al. (2024). The main fuels for GFR reactor burn-up analysis are UN-PuN (Uranium-Plutonium Nitride) and MA fuel rods. The number of fuel rods is arranged on 127 hexagonal lattice assemblies. The fuel configuration uses a heterogeneous core consisting of 5 fuel variations, namely F1, F2, F3, F4, and F5. Each fraction has a different composition of Pu including F1 = 8%, F2 = 9%, F3 = 10%, F4 = 11%, F5 = 12%. The fuel ring configuration for five fuel variations is F1 = 3 rings, F2 = 1 ring, F3 = 1 ring, F4

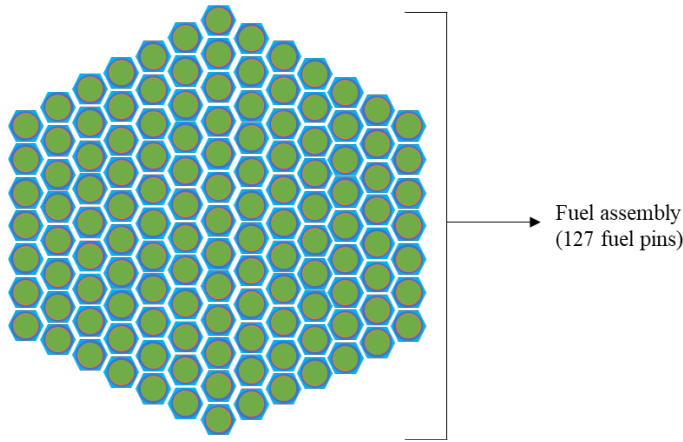


Figure 2. Fuel Pin Configuration on Fuel Assembly

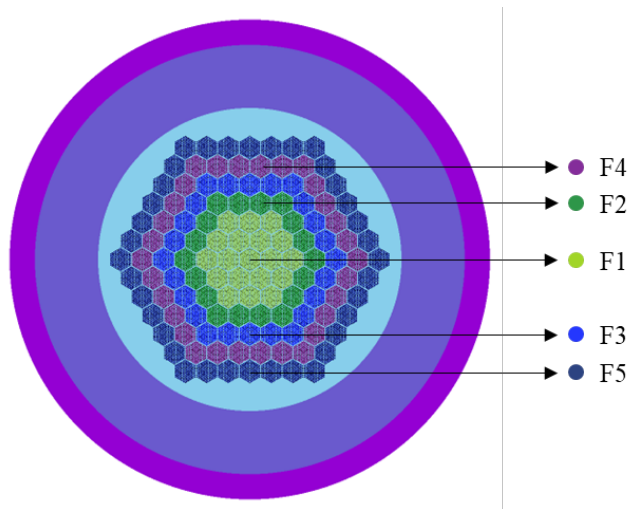


Figure 3. Heterogeneous Reactor Core Configuration with 5 Fuel Variations (F1, F2, F3, F4, F5)

= 1 ring, F5 = 1 ring. The placement of the MA fuel rods is adjusted to the very high flux point at the reactor core. Two MA fuel materials, Neptunium (Np) and Americium (Am), were utilized. This study used 6 MA fuel rods placed in a fuel assembly with a very high neutron flux value.

2.2 Preparation of Material Calculations with OpenMC

Calculation of GFR fuel material using Monte Carlo-based neutron transport method. Neutron transport relates to the rate of change or population of neutrons within a reactor. The rate of change of neutrons is influenced by several factors: neutron leakage, absorption reactions, neutron sources from fission reactions, and scattering reactions. Neutron transport is derived from the neutron diffusion Equation (1) as follows:

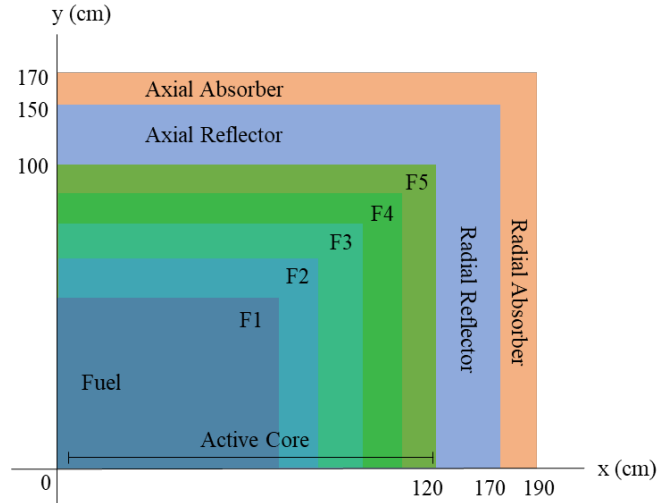


Figure 4. Quarter-Core Reactor Configuration Axially with Five Fuel Variations

Table 1. Material Parameters and Specifications

Parameters	Specifications
Main fuel material	UN-PuN
Minor actinide material	Np dan Am
Fuel/Gap/Cladding/Coolant volume fraction	60%/0.5%/10%/29.5%
Gap & coolant material	Helium (He)
Cladding material	SiC (Silicon Carbide)
Pin pitch	1.45 cm
Assembly pitch	17.04 cm
Number of fuel rings	7
Number of fuel assemblies	127
Number of MA fuel rods	6
Reactor core height	120 cm
Reactor core wide	200 cm
Reactor core configuration	Heterogeneous 5 fuel variations
Lattice shape	Hexagonal prism

$$\left[\frac{1}{v} \frac{\partial}{\partial t} + \Omega \cdot \nabla + \Sigma(r, E, t) \right] \Phi(r, \Omega, E, t) = \int_0^\infty dE' \int_{4\pi} d\Omega' \Sigma_s(r, \Omega', \cdot, \Omega, E', E) \Phi(r, \Omega', E', t) + \frac{X(E)}{4\pi} + \int_0^\infty dE' \int_{4\pi} d\Omega' v(E') \Sigma_f(r, E', t) \Phi(r, \Omega', E', t) + Q(r, \Omega, E, t) \tag{1}$$

If it is assumed to be in a steady state or independent of time, then the neutron transport Equation (1) becomes Equation (3)

Table 2. The Fuel Rod Indicator Data and Neutron Flux Value

Indicator	Neutron flux for F1 (neutron/cm.s)	Neutron flux for F2 (neutron/cm.s)	Neutron flux for F3 (neutron/cm.s)	Neutron flux for F4 (neutron/cm.s)	Neutron flux for F5 (neutron/cm.s)
G1	4.85E+13	4.62E+13	4.41E+13	4.28E+13	4.05E+13
F1	7.69E+13	7.34E+13	7.01E+13	6.74E+13	6.39E+13
E1	1.02E+14	9.75E+13	9.30E+13	9.00E+13	8.53E+13
D1	1.24E+14	1.18E+14	1.13E+14	1.09E+14	1.04E+14
C1	1.41E+14	1.34E+14	1.27E+14	1.24E+14	1.17E+14
B1	1.51E+14	1.43E+14	1.37E+14	1.33E+14	1.26E+14
A1	1.54E+14	1.47E+14	1.40E+14	1.36E+14	1.28E+14
B4	1.51E+14	1.44E+14	1.37E+14	1.33E+14	1.25E+14
C7	1.41E+14	1.34E+14	1.28E+14	1.24E+14	1.17E+14
D10	1.24E+14	1.18E+14	1.13E+14	1.09E+14	1.03E+14
E13	1.03E+14	9.77E+13	9.30E+13	9.02E+13	8.50E+13
F16	7.70E+13	7.32E+13	6.99E+13	6.75E+13	6.33E+13
G19	4.86E+13	4.63E+13	4.44E+13	4.25E+13	4.01E+13

as follows:

$$\lim_{t \rightarrow \infty} \Phi(r, \Omega, E, t) = 0 \tag{2}$$

$$\begin{aligned} [\Omega \cdot \nabla + \Sigma(r, E, \Omega)] \Phi(r, \Omega, E, t) &= \int_0^\infty dE' \int_{4\pi} d\Omega' \Sigma_s \\ (r, \Omega', \Omega, E', E) \Phi(r, \Omega', E') &+ \frac{X(E)}{4\pi} + \int_0^\infty dE' \int_{4\pi} \\ d\Omega' v(E') \Sigma_f(r, E', t) \Phi(r, \Omega', E') &+ Q(r, \Omega, E) \end{aligned} \tag{3}$$

The distribution of steady-state sources or not time-dependent Q, r, Ω, E . now includes the initial state (Romano et al., 2015).

The neutron transport method is used in OpenMC 0.14.1 programs. This program has complete features for neuronal analysis needs in cases such as entire core. OpenMC has the advantage of being open-source (unlicensed) and using Python, making it easier for users to learn. The material used for calculation is written in the input script code in the form of material.xml. Other OpenMC input files needed to produce more comprehensive calculations are geometry.xml, setting.xml, tallies.xml, and plot.xml. Nuclear library data for cross-section material using type ENDF/B-VII.1. The depletion chain used in the ENDF/B-VII.1 library is fast spectrum type version 0.12.

2.3 Determination of Material Parameters and Specifications

Material parameters and specifications are required as input data material.xml in the OpenMC program described in Table 1. Using six MA fuel rods is good enough to reduce neutron flux values and power peaking for each fuel assembly (Syarifah et al., 2024). The primary fuel arranged in the reactor core is coated with reflectors and absorbers. The reflector uses SiC (Silicon Carbide) material, and the absorber uses B₄C (Boron Carbide) material. SiC has a percentage level of albedo (the number of neutrons reflected by the reflector surface) up to

85.17% (Rafliis et al., 2021), and B₄C has a reasonable neutron leakage absorption rate (Harsanti, 2010). The design of the fuel pin, fuel assembly, and reactor core is shown in Figures 1-4.

The fuel assembly is a group of several fuel pins arranged to form a hexagonal prism of lattice. The active reactor area or core consists of several configurations of fuel assembly F1, F2, F3, F4, and F5 arranged to form a large lattice hexagonal prism. The total pins for each fuel assembly configuration include 19 fuel pins of F1, 18 fuel pins of F2, 24 fuel pins of F3, 30 fuel pins of F4, and 36 fuel pins of F5.

2.4 Neutron Flux Analysis to Determine MA Fuel Rods Placement

A suitable reactor core design will produce an even distribution of neutron flux or flattening power. Generally, high flux values are in the central region of the reactor (F1, F2, F3). If visualized in the distribution of color distribution, the central region of the reactor will be solid red. This shows that there is peaking power in the central area of the reactor. Therefore, it is necessary to add MA fuel rods to reduce the neutron flux value in the F1, F2, and F3 regions so that the power produced is more evenly distributed. MA fuel rods consist of Neptunium (Np) and Americium (Am), which are optimized using four designs. The configuration of the four designs is shown in Figure 5.

The MA fuel rod contains 100% Np and Am material, so it is expected to reduce flux distribution and maintain the stability of reactor criticality for each burn-up point. These four MA fuel rod configurations are used to analyze fission product waste generated from burn-up for five years or 1825 days. Fission product waste produced can be in the form of radioisotope or radiopharmaceutical.

2.5 Reactor Criticality Analysis

Reactor criticality can be reviewed under three conditions, including sub-critical, critical, and super-critical. Sub-critical

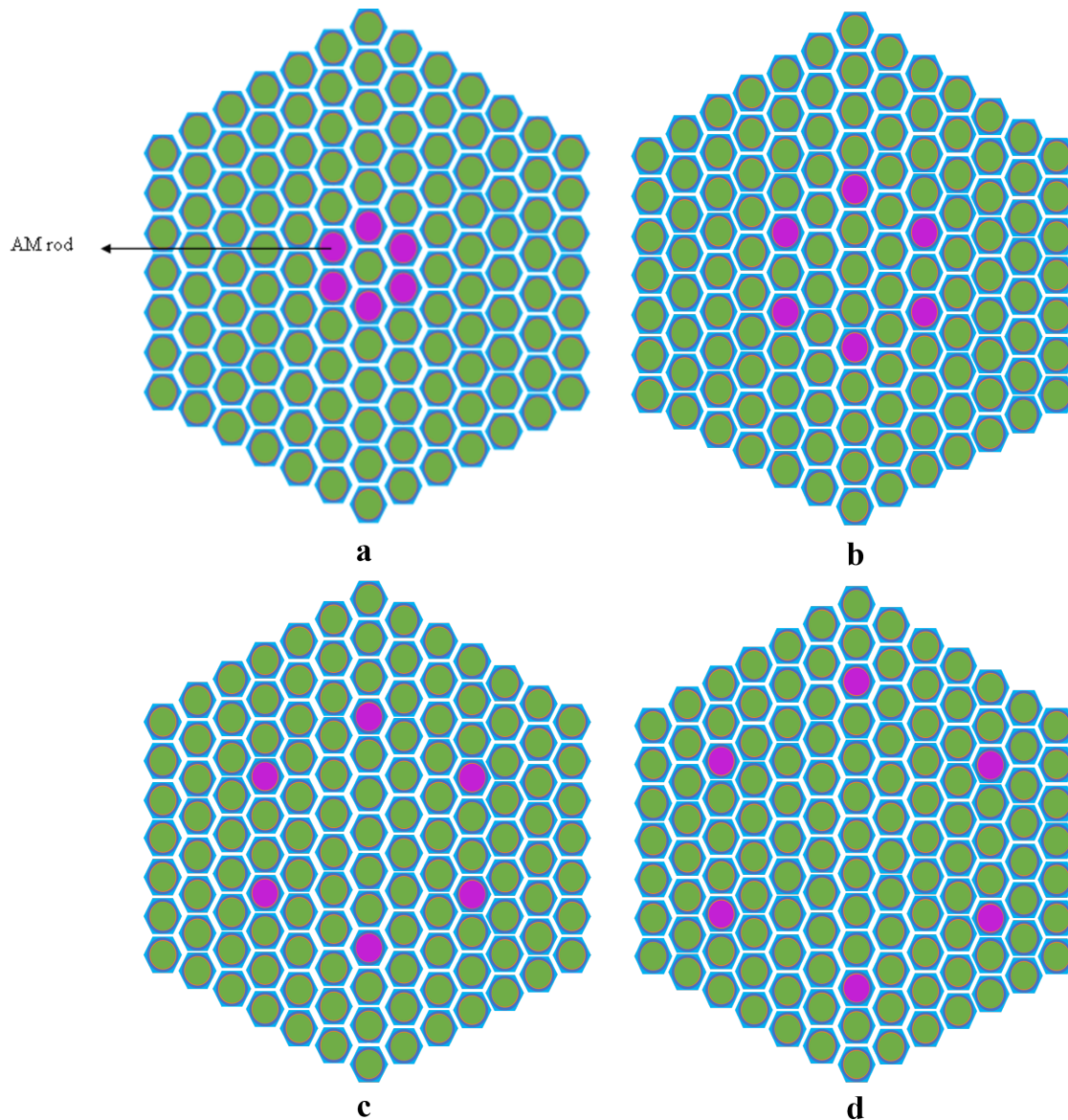


Figure 5. Configure the Placement of MA Fuel Rods in the Fuel Assembly for (a) Design 1, (b) Design 2, (c) Design 3, and (d) Design 4

conditions where the reactor has a k_{eff} value < 1 , critical conditions have a k_{eff} value $= 1$, and super-critical have a k_{eff} value > 1 . Adding MA fuel rods can reduce the k_{eff} value of the heterogeneous core design 5 fuel variations close to critical conditions ($k_{eff} \approx 1$) or have an excess reactivity value of $< 1\%$ until the end of burn-up. Mathematically, the k_{eff} value or the value of the effective multiplication factor and excess reactivity ($\Delta k/k$) can be written as the following Equations (4 & 5).

$$k_{eff} = \epsilon L_f \rho L_t f \eta \tag{4}$$

$$\frac{\Delta k}{k} = \left(\frac{k_{eff} - 1}{k_{eff}} \right) \times 100\% \tag{5}$$

Where ϵ is the fast fission factor, L_f is the probability of fast non-leakage, ρ is the resonance capture probability, L_t is the probability of thermal non-leakage, f is the thermal utilization factor, and η is the thermal utilization factor.

2.6 Fission Product Waste Analysis in the Form of Radio-pharmaceuticals

The last step in this study is to analyze the waste from burning MA fuel rods placed in the fuel assembly. The burn-up results of MA fuel rods can be viewed from the change in mass (kg) from the beginning of burn-up (0th year) to the end of burn-up (5th year). In addition, new isotopes appear due to the decay of MA fuel rods. The new isotope is a waste fission product with

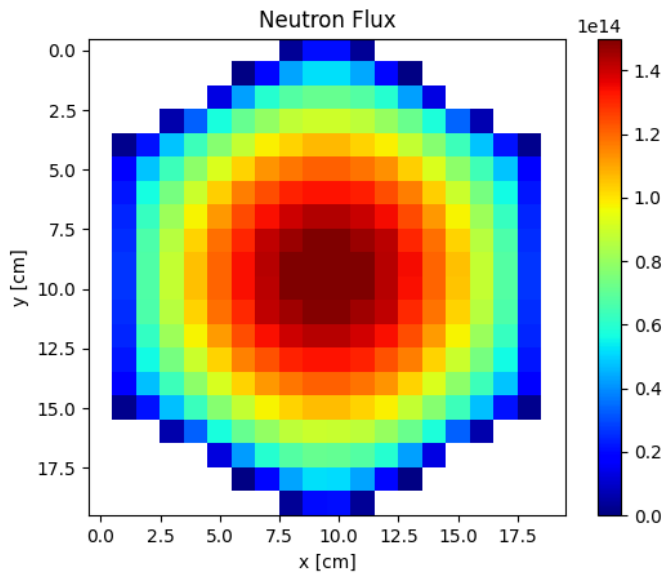


Figure 6. Neutron Flux Distribution for One Fuel Assembly

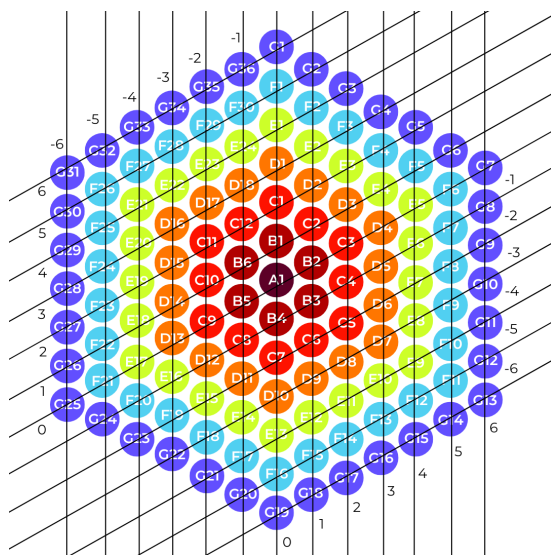


Figure 7. The Fuel Rod Indicator for One Fuel Assembly

a shorter $T/2$ decay rate period, so it is more environmentally friendly and can be used for pharmaceutical or medical needs. The waste is then reviewed, and the change in mass (kg) from the beginning to the end of burn-up is noted.

3. RESULTS AND DISCUSSION

3.1 Neutron Flux for One Fuel Assembly

The result of the neutron flux (neutron/cm.s) calculation for the xy-axis radial cross-section is shown by the distribution of color distribution in Figure 6. The highest flux value is in the middle area of the fuel assembly, which is indicated by the distribution of solid red. Neutrons that cross a unit of volume in units of time in the middle region have a more significant

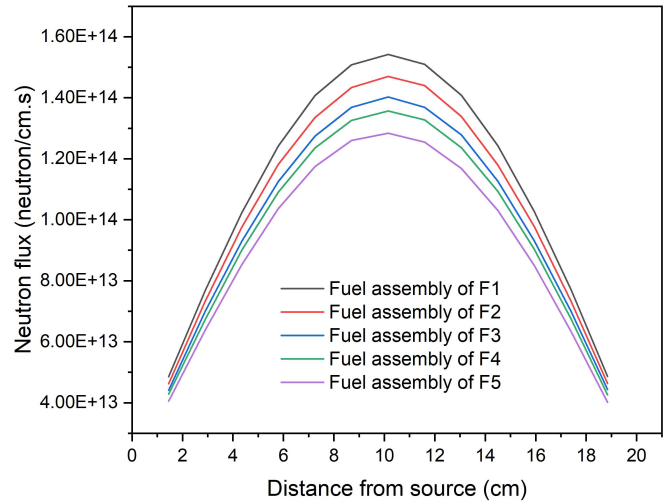


Figure 8. Graph of Neutron Flux Values

number than in the periphery. If viewed using five types of fuel assembly configurations F1, F2, F3, F4, and F5 for one dimension (x or y axis direction), the same graphic form is obtained, namely peaking flux in the center area of the fuel assembly. The fuel assembly of F1 has a higher neutron flux value than others because it uses the primary fuel material (UN) with a large percentage of as much as 92%. Meanwhile, the fuel assembly of F5 only uses 88% UN, so the flux value is small (shown in Table 2). The fuel rods indicator and the distribution of flux values for one reactor dimension are shown in Figures 7 & 8.

The results of neutron flux identification can serve as a reference for positioning the MA fuel rods within the fuel assembly configuration for a full core design. Four designs for MA fuel rod placement were utilized, each depending on the reference flux value of the respective fuel rod (see Figure 5). MA fuel rods were allocated to areas with high flux values to achieve a flatter neutron flux profile and approach a near-critical value ($k_{eff} \approx 1$). As depicted in Figure 3, MA fuel rods were positioned in regions F1, F2, and F3.

3.2 Criticality Value after MA Fuel Rods Addition

Before adding MA fuel rods, the reactor criticality value for heterogeneous core configurations of 5 fuel variations was 1.0386 at the beginning and 1.0172 at the end of burn-up. This shows that the decrease in the value of k_{eff} is very significant until the end of burn-up, which is $\pm 2\%$, and the excess reactivity value is still more than 1% (Prasetya et al., 2024). Adding MA fuel rods can reduce the k_{eff} value to near critical condition ($k_{eff} \approx 1$) and reach an excess reactivity value of $< 1\%$. In addition, adding MA fuel rods can provide stability in decreasing the value of k_{eff} so that it does not experience a significant decrease for each burn-up point. The results of reactor criticality calculations for heterogeneous core configurations of 5 fuel variations for each MA fuel rod placement design are shown in Figure 9.

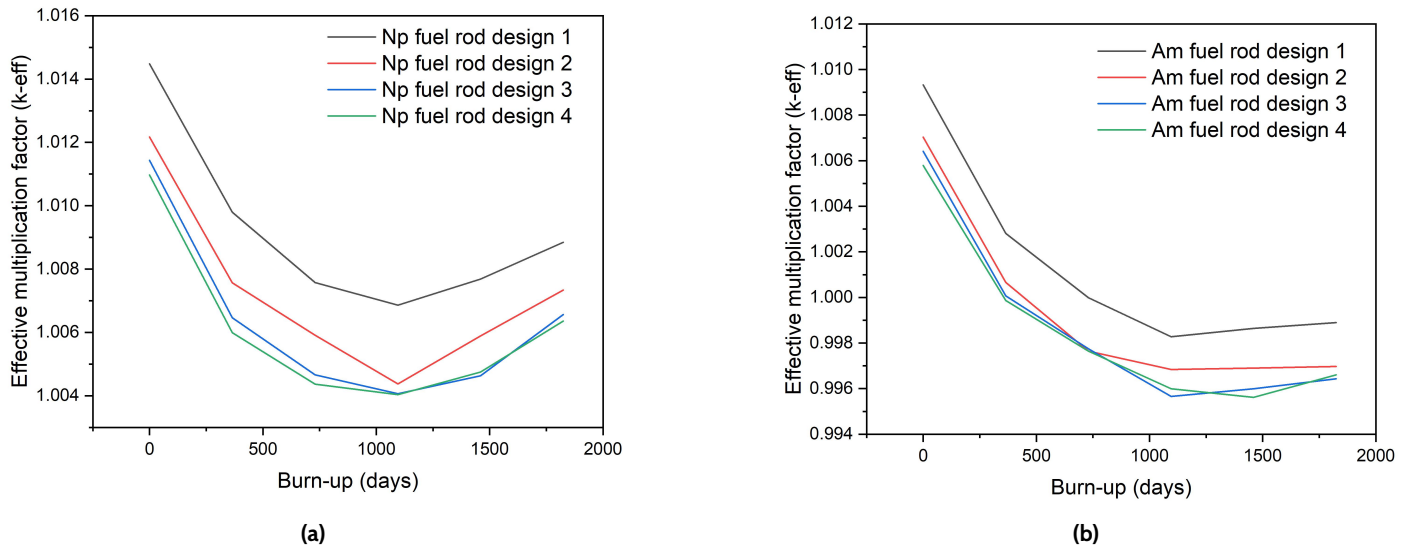


Figure 9. Reactor Criticality Value of Adding MA Fuel Rods for Material (a) Np (b) Am

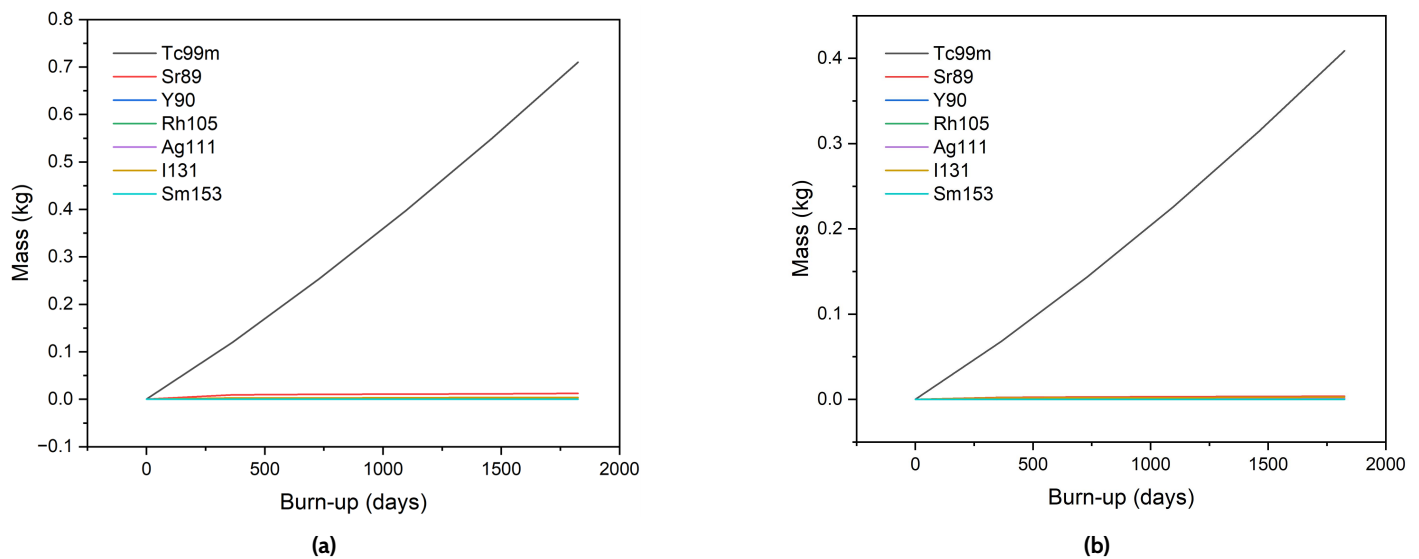


Figure 10. Production of Radiopharmaceutical Waste from Burn-up MA Fuel Rod for Material (a) Np Design 4 (b) Am Design 1

The graph in Figure 9 shows a slight increase in the value of k_{eff} at the burn-up point of 3rd year or 1095 days for Np and Am materials. This is because the fertile material from the primary fuel reaches the breeding process and decays into fissile material. MA materials in the form of Np237, Am241, and Am243 will maintain the fission reaction longer and cause criticality to increase with the burn-up process. The addition of Np fuel rod material design 4 gets the smallest excess reactivity value when compared to other designs, reaching 0.63% at the end of burn-up. Meanwhile, adding Am fuel rod material for all designs gets a criticality value that tends to be sub-critical at the end of burn-up so that the excess reactivity value becomes negative. The addition of Am fuel rods to design 1 is the best

design, because the criticality value above the critical point ($k_{eff} > 1$) can be maintained longer than other AM rod designs.

3.3 Analysis of Radiopharmaceutical Material from MA Fuel Rod Burn-up

Np fuel rod design 4 and Am fuel rod design 1 are used for further calculations related to fission product analysis for the probability of the emergence of new waste material in the form of radiopharmaceuticals. The radiopharmaceutical materials analyzed for this study were Tc99, Sr89, Y90, Rh105, Ag111, I231, and Sm153. Radiopharmaceutical materials are generally used for medical needs, so the decay time or T/2 is very short in periods of days, hours, minutes, and sec-

onds. Technetium-99 or Tc99 radiopharmaceutical material is the main product of fission decay U235 and minor actinides Np237, Am241, and Am243 (Rohadi, 2011). This makes it the most available and commonly used technetium isotope in nuclear medical requirements. Meanwhile, Sr89, Y90, Rh105, Ag111, I231, and Sm153 are fission products with a small probability of producing radiopharmaceutical material waste. The production of radiopharmaceutical material waste from the burn-up of MA fuel rod is shown in Figure 10 and Table 3.

Table 3. Mass of Radiopharmaceutical Waste

MA Fuel Rod	Radiopharmaceutical Isotope	Final Burn-up Mass (kg)
Np Design 4	Tc99m	0.71
	Sr89	0.00123
	Y90	0.000081
	Rh105	0.00057
	Ag111	0.00017
	I131	0.0038
Am Design 1	Sm153	0.00011
	Tc99m	0.41
	Sr89	0.0038
	Y90	0.000023
	Rh105	0.00068
	Ag111	0.00086
	I131	0.0024
	Sm153	0.00015

Based on Figure 10, Tc99m material has the highest probability of producing radiopharmaceutical waste with a final burn-up mass of 0.71 kg for Np material and 0.41 kg of Am material. Other radiopharmaceutical waste materials such as Sr89, Y90, Rh105, Ag111, I231, and Sm153 have masses that tend to be very small (less than the order of 10^{-3} kg). It can be said that the burn-up of an MA fuel rod can produce radiopharmaceutical waste that has a mass of isotopes that are very little or less than 1 kg. The process plays a role in reducing the negative impact of MA waste on the environment and has a positive effect by producing radiopharmaceutical waste that is useful in the medical field.

4. CONCLUSIONS

The use of minor actinide materials (MA) Np and Am is very helpful in flatten the neutron flux values. The placement of the MA fuel rod in this study is based on the highest flux value in the middle area of the fuel assembly. Referring to the results of the flux value, 4 MA fuel rod placement designs were obtained to reduce the flux value and reactor criticality to near critical ($k_{eff} \approx 1$). Np fuel rod design 4 and Am fuel rod design 1 get the best criticality score among other designs. However, all Am fuel rod designs tend to be sub-critical at the end of burn-up. The burn-up of Np and Am produces Tc99m radiopharmaceutical waste with more mass when compared to Sr89, Y90, Rh105, Ag111, I231, and Sm153. The probability of radiopharmaceutical

waste production for this reactor research is very small or less than 1 kg. Using Np and Am materials in a reactor design can effectively lower neutron flux values and produce a more even flux distribution. Placing MA fuel rods based on the highest flux value in the middle area of the fuel assembly can optimize the flux value and reactor criticality. However, the burn-up of Np and Am produces radiopharmaceutical waste with a higher mass compared to other isotopes, and the probability of radiopharmaceutical waste production for this reactor design is very low.

5. ACKNOWLEDGMENT

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