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Utilising ^{241}Am as burnable poison in proliferation resistant PWR

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ABSTRACT

The increased need for energy, as well as the necessity for energy-intensive solutions to tackle climate change, has increased interest in nuclear power generating as a low-carbon energy source. While nuclear energy offers substantial benefits in reducing greenhouse gas emissions, it also raises concerns regarding nuclear proliferation. In this study, the viability of utilising nuclear proliferation-resistant fuel in a PWRs without the need for an additional burnable absorber was assessed by integrating ^{241}Am into the fuel composition. When ^{241}Am was used as a burnable absorber instead of IFBAs, the potential changes in reactivity feedback parameters, peaking factors, and power profiles were investigated. The influence of ^{241}Am-doped fuel on the cycle's duration and the shut-down margin was also explored. It is shown that using ^{241}Am-doped fuel can enable a PWR to operate safely and reliably within its design boundaries. Moreover, it offers a proliferation-resistant fuel cycle without requiring any additional burnable absorbers.

1. Introduction

The profound risks associated with the uncontrolled spread of nuclear materials and technology are long recognised by the international community (Burt, 1977). At the heart of this concern is the prevention of states, particularly those perceived to have dubious intentions, from being endowed with nuclear weapon capabilities. Through a myriad of international agreements, with the Nuclear Non-Proliferation Treaty (NPT) being most noteworthy, combined with consistent intelligence collaboration and diplomatic interventions, efforts are made to alleviate the potential dangers presented by nuclear weapons (Bunn, 2003). The primary objective is not only to diminish the likelihood of grave nuclear confrontations but also to maintain the wider global structure.

In this context, a robust non-proliferation ethos is established for several reasons. Besides the urgent necessity to reduce the chances of devastating wars, this ethos is seen as essential in facilitating diplomatic solutions to enduring conflicts. As a result, a sense of regional stability is promoted, leading to the fostering of trust between nations. Moreover, the principle of non-proliferation is considered vital for safeguarding the welfare of current and future generations and protecting human societies and the greater environment from the prolonged consequences of nuclear fallout and pollution.

In fuel utilisation of spent nuclear fuel for making nuclear weapons, two main techniques are proposed. The first, production of plutonium in a dedicate reactor with the intention to reach appropriate plutonium quality. The other method is believed to work through altered durations of fuel exposure within the typical power reactor. Even though this method might produce weapons-grade plutonium, significant interruptions to the reactor's regular functioning would be required (Ronen and Kimhi, 1991). It should be stressed that weapons-grade plutonium is not found in the composition of the spent fuel from a common power reactor at the cycle's end.

The classification of plutonium as weapon-grade is determined by its composition. It is mandated that, for plutonium to be considered weapon-grade, the presence of the ^{239}Pu isotope must be at least 93 wt %, and the ^{240}Pu isotope should not exceed 7 wt%. When it comes to typical Light Water Reactors (LWRs), the isotopes ^{239}Pu and ^{240}Pu make up roughly 57 wt% and 26 wt%, respectively, of the entire plutonium content found when the fuel is discharged (Aghara and Beard, 2002), thus deeming it unsuitable for weaponization. For the isotope ^{238}Pu, a range from 1 % to 3 % of the total plutonium content is noted upon the fuel being released (Ronen et al., 2010).

The fuel is deemed proliferation-resistant due to the properties of ^{238}Pu when the ratio of ^{238}Pu to total plutonium (hereafter called ^{238}Pu/TotPu) equals or exceeds 6.00 wt% (Kessler, 2007). ^{238}Pu originates from the β-decay of ^{238}Np, which results from the neutron absorption by the ^{237}Np isotope. Furthermore, another source of ^{238}Pu is ^{241}Am. After capturing a neutron, ^{241}Am transforms into ^{242}Am, which has a half-life

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Table 1
Design parameters and operation limits used in the simulations (Ames Ii et al., 2010; DiGiovine and Gheorghiu, 1999; Duke Energy, 2018; U.S.NRC, 1982).

Reactor Type	3-Loop PWR
Coolant inlet temperature at full power (°C)	286.0
Coolant outlet temperature at full power (°C)	326.6
Average fuel temperature at full power (K)	820.5
Power Output (MWt/MWe)	2900/965
Operation power load (%)	100
System pressure (MPa)	15.5
Control rod material	Ag - In - Cd
Number of assemblies	157
Rod array	17 × 17
Assembly pin pitch (cm)	1.26
Fuel pellet radius (cm)	0.410
IFBA thickness (cm)	0.00256
B-10 loading (Mg/cm)	0.772
Number of control rods/guide tube	24/1
Number of BA rods	24
Fuel assembly pitch (cm)	21.50
Fuel assembly height (cm)	365.76
Cladding material	Zircaloy-4
UO_2 fuel density (% of TD)	95
Nuclear Enthalpy Rise Hot Channel Factor ($F_{\Delta H}$)	≤1.66
Heat Flux Hot Channel Factor (F_Q)	≤2.41
Moderator temperature coefficient (pcm/°F)	$-50 \leq MTC < 0$
Shutdown Margin (pcm)	≤ -1770

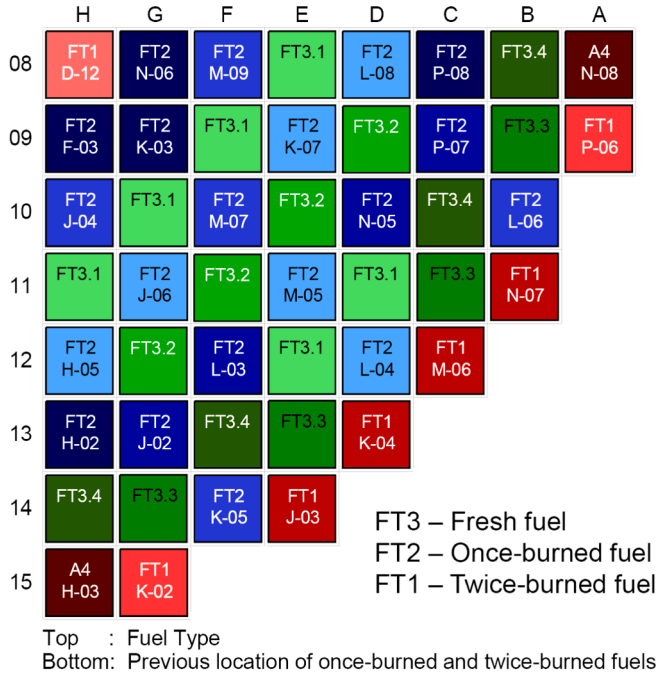

Fig. 1. Fuel loading pattern (Amjad et al., 2014).

of 16 h and undergoes β decay to become ^{242}Cm with a half-life of 163 days; eventually, ^{238}Pu is produced through α decay (Ronen et al., 2012).

A research by Ronen et al. in 2010 (Ronen et al., 2010) delved into the potential of ^{241}Am to act as a proliferation-resistant burnable poison within PWR. Their findings showed that by incorporating ^{241}Am, the $^{238}Pu/Pu$ ratio in the expelled fuel noticeably increases, enhancing its anti-proliferation characteristics. This inclusion allows for significant decreases in reactivity control mechanisms without considerably shortening the fuel cycle length since ^{241}Am acts as a burnable absorber. They also highlighted that while ^{237}Np is another additive suggested for boosting proliferation resistance, the lesser reduction in fuel cycle duration and greater burnable poison savings rendered by ^{241}Am make it a more beneficial choice.

For the safe and reliable operations of nuclear reactors, control of reactivity is essential. During reactor operation of a typical PWR, control over reactivity is maintained by the use of Burnable Absorbers (BAs). Among them, the Integral Fuel Burnable Absorbers (IFBAs), developed by Westinghouse Electric Company, are commonly used (Alameri and Alrwashdeh, 2021). IFBAs are obtained by coating the outer surfaces of fuel pellets with a thin layer of ZrB_2 (Franceschini and Petrović, 2009). Depending on the type of fuel used and the level of uranium enrichment, the number of fuel rods containing IFBA and the thickness of the IFBA coating varies (Alameri and Alrwashdeh, 2021).

In their study, (Bolukbasi and Margulis, 2024) examined the effects of ^{241}Am-doped fuel on PWR operations. The research revealed that, in order to achieve a 6.00 wt% $^{238}Pu/TotPu$ ratio in the fuel composition, a minimum ^{241}Am ratio of 0.112 wt% was required. Furthermore, it was noted that the use of ^{241}Am resulted in an important reduction in reactivity. The study demonstrated that by incorporating ^{241}Am into the fuel composition, it is possible to achieve a proliferation-resistant fuel cycle within the operational and design limits of a PWR.

This study serves as a continuation of the previous research, with the primary objectives being the establishment of a proliferation-resistant fuel cycle and the elimination of the necessity for additional burnable absorbers, such as IFBA, which are complex and costly to implement. The influence of the ^{241}Am-doped fuel on the fuel cycle length, reactivity feedback parameters, peaking factors, shutdown margin and power profiles in a typical PWR operation were assessed using the CASMO4/SIMULATE3 code package.

2. Materials and methods

The methodology employed in this study is built upon the methodologies of previous studies in the literature. In the simulations, the CASMO4/SIMULATE3 software package was used with the ENDF/B-VI neutron data library as in prior research (Bolukbasi and Margulis, 2024). A Westinghouse Electric Company standard 3-loop PWR reactor NPP was utilized, and certain simulation parameters and characteristics of the NPP are presented in Table 1.

Two distinct scenarios were devised within the study. One scenario was constructed as a PWR operation with only IFBA utilized in the fuel assembly, while in the other scenario, only ^{241}Am was employed as the burnable absorber (BA).

Fig. A1 displays the initial cycle fuel loading pattern utilized in the simulations. The cross-sections of Pyrex rods used in the initial cycles (see Fig. A2) and their distributions within the fuel assembly (see Fig. A3) were employed in accordance with the previous study (Bolukbasi and Margulis, 2024). In the scenario employing IFBA, IFBA loading was applied to fresh fuel assemblies beginning from the second cycle, and the distribution of fuel rods containing IFBA within the fuel assembly is illustrated in Fig. A4.

Every fuel assembly introduced into the reactor was individually tracked in the simulations. It was observed that when the percentage of ^{241}Am used in later cycles was implemented in the initial cycle, certain fuel assemblies from that first cycle failed to attain a $^{238}Pu/TotPu$ ratio exceeding 6.00 wt% during the subsequent second and third cycles. As a result, varied percentages of ^{241}Am were selected for the fuels used in the first cycle.

To achieve a more uniform axial power distribution, a 15.24 cm IFBA-free zone at both the top and bottom sections of the fuel assembly was chosen. Axial blankets were not incorporated into the fuel rods. Furthermore, when determining the $^{238}Pu/TotPu$ ratio, ^{242}Cm, which possesses a half-life of 163 days and transforms into ^{238}Pu via alpha emission, was factored into the ^{238}Pu quantity (Ronen et al., 2010).

All simulations were undertaken with all control rods fully withdrawn, operating under the hot full power (HFP) conditions. Under these conditions, the reactor runs at its maximum power, and the temperatures of the coolant and fuel align with those specified in Table 1. In

Table 2
Fuel compositions, number of fuel assemblies, and core position of the fresh fuels.

Number of fresh FAs	Uranium enrichment (wt.%)	Core position	^{241}Am level (wt.%) in subsequent cycles (where ^{241}Am-doped fuel utilised)	Number of IFBA rods in subsequent cycles (where ^{241}Am-free fuel utilised)
20	4.75	FT.3.1	0.280	112
16	4.80	FT.3.2	0.280	112
16	4.85	FT.3.3	0.210	112
12	4.90	FT.3.4	0.200	112

both scenarios, from the second cycle onwards, the fuel loading pattern shown in Fig. 1 and the fuel compositions detailed in Table 2 were utilised. According to this loading pattern, 64 fresh fuel assemblies are introduced into the reactor for each cycle, and 35 once-used and 29 twice-used fuel assemblies are removed. Furthermore, to effectively address power peaks, fresh fuels were divided into four distinct categories.

It is crucial that the fuel compositions comply with the reactor's operational and safety boundaries. As such, emphasis was placed on peaking factors, including the nuclear enthalpy rise hot channel and heat flux hot channel factors. Additionally, reactivity feedback parameters, such as boron coefficient (BC), uniform Doppler coefficient (UDC), isothermal temperature coefficient (ITC), and moderator temperature coefficient (MTC) were analysed. Finally, the Average axial relative power distributions of the fuel models, comprising 12 axial nodes, and their assembly-wise 2D relative power fractions were evaluated.

The shutdown margin stands as a critical parameter in ensuring the safe and dependable operation of a Nuclear Power Plant (NPP). To assess the potential ramifications of utilizing ^{241}Am-doped fuel on the shutdown margin, calculations were carried out using the methods outlined in (Bolukbasi and Margulis, 2024).

3. Results and discussion

3.1. Neutronic analysis

In Fig. 2, curves representing the kinf values for various fuel models are illustrated. These models include a fuel model both free of BA and ^{241}Am and enriched at 4.45 wt%, a fuel model devoid of ^{241}Am but containing 112 IFBA rods, with a boron loading of 0.772 ^{10}B/cm, and fuel models with varying levels of ^{241}Am between 0.100 and 0.280 wt%.

In all depicted curves, a swift reduction in the kinf values at the beginning of the fuel models' life is noticeable. This reduction trend occurred as a result of the rapid generation of xenon-135 and samarium-149 fission products. Possessing high thermal neutron capture cross-sections, these fission products affect the neutron economy negatively prior to achieving equilibrium (Attom et al., 2019; Galahom, 2016).

From Fig. 2, it is observed that the inclusion of IFBA or ^{241}Am in the fuel composition predictably imposes a suppression on reactivity at the beginning of the fuel's life cycle, leading to a decline in the k_{inf} value. In addition, this reduction increases with the increase in ^{241}Am concentration in fuel composition. Finally, the reactivity swing associated with the IFBA presence disappears when ^{241}Am is used and a more traditional behaviour of the reactivity curve is observed.

In ^{241}Am-doped fuel models, a gradual declining trend in k_{inf} values throughout the cycle is observed. Despite this, fuel models with relatively high concentrations of ^{241}Am at 0.220 wt% and 0.280 wt% display a small horizontal trend up to approximately 3.00 MWd/kgU. Subsequently, these models equalize with the reference model at around 49.00 MWd/kgU, before exhibiting higher k_{inf} values than the reference fuel model during the rest of irradiation period. For example, a k_{inf} value approximately 0.32 % (281 pcm) higher than the reference fuel model is owned by a fuel model doped with 0.100 wt% ^{241}Am at 60.00 MWd/kgU. Meanwhile, a k_{inf} value that is approximately 0.81 % (714 pcm) higher is possessed by a fuel model doped with 0.280 wt% ^{241}Am at 60.00 MWd/kgU.

In Fig. 3a and b, the ratios of ^{238}Pu/TotPu and ^{240}Pu/TotPu for the fuel models, discussed in Fig. 2, are provided. Upon examination of Fig. 3a, it is seen that the use of ^{241}Am in the fuel model causes a peak in the ^{238}Pu/TotPu ratio at the beginning of the life of all fuel-dopped models. The peak in the ^{238}Pu/TotPu ratio at the beginning of the fuel life increases with the rise in the concentration of ^{241}Am within the fuel composition. For example, in a fuel model doped with 0.100 wt% ^{241}Am, the ^{238}Pu/TotPu ratio is 17.14 %, whereas in fuel models doped with 0.112 wt%, 0.220 wt%, and 0.280 wt% ^{241}Am, the ratios are 18.69 %, 29.65 %, and 34.00 % respectively. These ratios in the fuel models are observed to decline to lower levels as burnup progresses, as the dominant reaction is conversion of ^{238}U to ^{239}Pu.

However, a point that must be noted is that in the fuel model doped with 0.110 wt% ^{241}Am, the ^{238}Pu/TotPu ratio falls below the 6.00 wt%

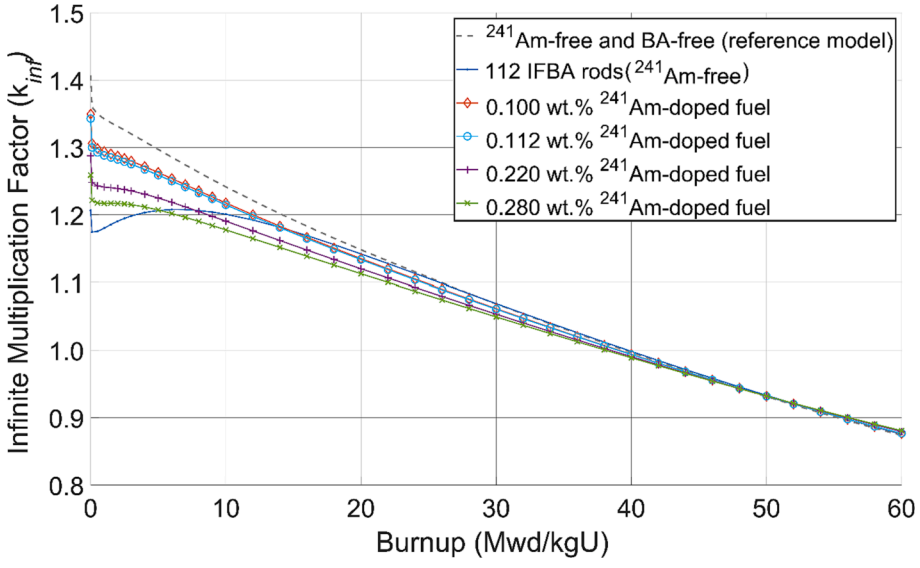

Fig. 2. Infinite multiplication factor (k_{inf}) for fuel assembly models with IFBA, and different ^{241}Am additions.

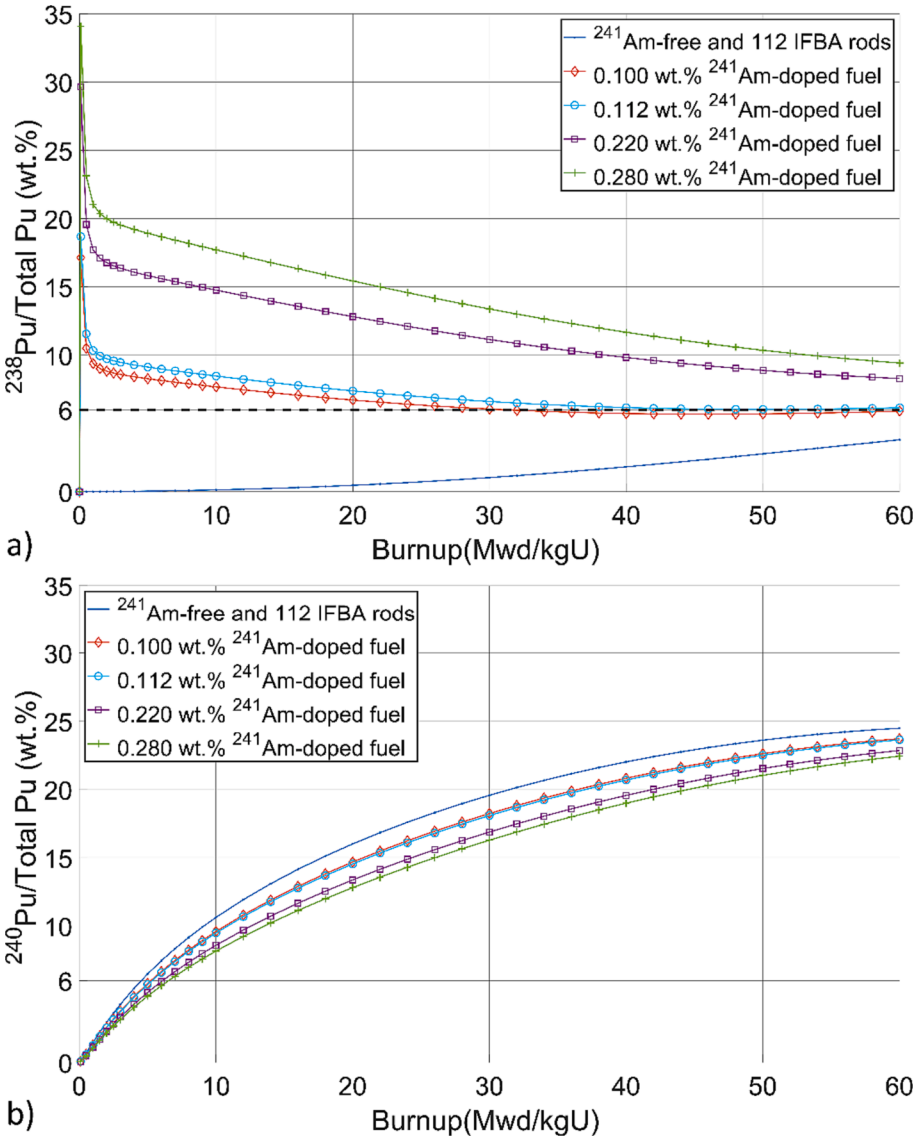

Fig. 3. ^{238}Pu/TotPu (a) and ^{240}Pu/TotPu (b) ratios for fuel assembly models with IFBA, and different ^{241}Am addition.

Table 3
Cycle parameters of the reference IFBA-equipped fuel cycle scenario.

Cycle No.	CBC- BOC (ppm)	CBC at 10 MWd/kgU (ppm)	BOC-k_{eff}	Maximum $F_{\Delta H}$	Maximum F_Q	Cycle Burnup (MWd/kgU)	Cycle EFPDs
1	1,514.8	940.1	1.16588	1.577	2.076	20.926	530.5
2	1,380.4	817.1	1.09462	1.646	1.922	18.237	462.3
3	1,823.8	1154.6	1.11548	1.651	2.008	20.710	525.0
4	1,726.0	1091.7	1.10950	1.615	1.942	20.300	514.6
5	1,747.2	1103.9	1.11068	1.624	1.958	20.375	516.5
6	1,743.6	1102.1	1.11047	1.622	1.954	20.363	516.3
7	1,744.2	1102.4	1.11050	1.623	1.956	20.365	516.3
8	1,744.1	1102.3	1.11050	1.623	1.956	20.365	516.3
9	1,744.1	1102.4	1.11050	1.623	1.956	20.365	516.3
10	1,744.1	1102.3	1.11050	1.623	1.957	20.365	516.3

level, which is considered the nuclear proliferation-resistant limit, at 31.00 MWd/kgU. The lowest level observed in this model is 5.74 wt% ^{238}Pu/TotPu at 46.00 MWd/kgU. On the other hand, in the fuel model doped with 0.112 wt% ^{241}Am, the lowest observed ^{238}Pu/TotPu ratio is 6.03 wt% at 50.00 MWd/kgU. Therefore, it can be said that the addition of 0.112 wt% ^{241}Am is the minimum amount required for maintaining a ^{238}Pu/TotPu ratio above 6.00 wt%, thus considered nuclear proliferation-resistant. Ronen et al. (2009) (Ronen et al., 2010) have indicated that this amount is 0.12 wt%. The discrepancy between these figures is undoubtedly due to the variations in the nuclear data libraries employed.

As observed in Fig. 3b, some variations are noted in the ^{240}Pu/TotPu ratios. For instance, at 30.00 MWd/kgU and 60.00 MWd/kgU, the ^{240}Pu/TotPu ratios for the ^{241}Am-free fuel model are identified as 19.58 and 24.49 wt% respectively. However, in the fuel model with the highest ^{241}Am content, which is 0.228 wt%, the ^{240}Pu/TotPu ratio declines to

Table 4
Cycle parameters of the ^{241}Am-doped fuel cycle scenario.

Cycle No.	CBC- BOC (ppm)	CBC at 10 MWd/kgU (ppm)	BOC-k_{eff}	Maximum $F_{\Delta H}$	Maximum F_Q	Cycle Burnup (MWd/kgU)	Cycle EFPDs
1	834.6	680.5	1.09941	1.598	2.186	19.320	489.8
2	1,678.7	739.9	1.11666	1.631	1.977	17.411	441.4
3	2,015.3	1010.3	1.12967	1.630	2.118	19.614	497.3
4	1,947.9	962.2	1.12523	1.602	2.041	19.266	488.4
5	1,963.9	973.4	1.12611	1.610	2.063	19.348	490.5
6	1,960.6	971.0	1.12592	1.608	2.058	19.329	490.0
7	1,961.5	971.6	1.12597	1.610	2.062	19.334	490.2
8	1,961.2	971.4	1.12596	1.610	2.061	19.333	490.1
9	1,961.3	971.5	1.12596	1.610	2.062	19.333	490.1
10	1,961.3	971.5	1.12596	1.610	2.061	19.333	490.1

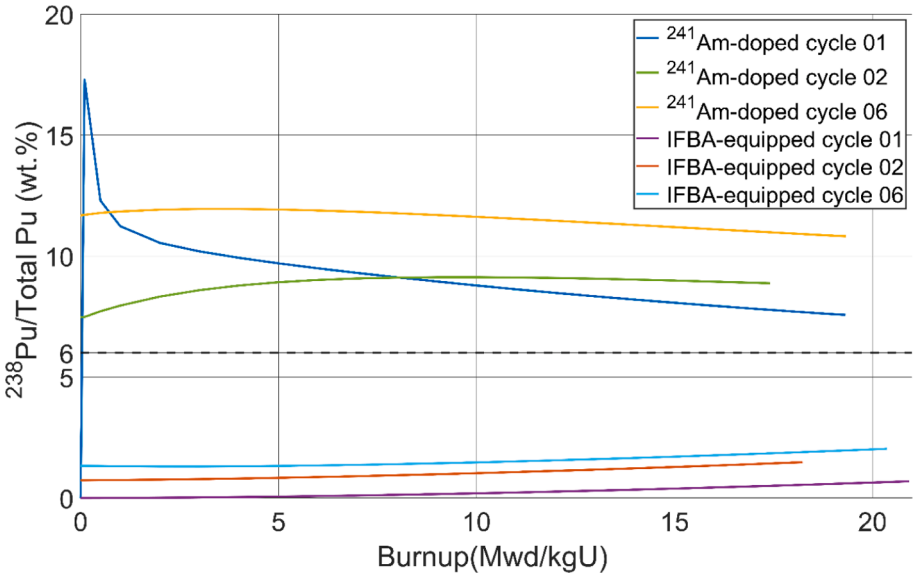

Fig. 4. Core average ^{238}Pu/TotPu ratios of cycles 1, 2, and 6 for both scenarios.

16.29 wt% at 30.00 MWd/kgU and further decreases to 22.45 wt% at 60.00 MWd/kgU. Therefore, it can be stated that significant alterations in the ^{240}Pu/TotPu ratio, a critical factor that complicates nuclear weapon production due to its tendency to undergo spontaneous fission (US The National Academy of Sciences, 1994), are not observed.

Lastly, in the fuel models doped with 0.220 wt% and 0.280 wt% ^{241}Am, a gradual decline in the ^{238}Pu/TotPu ratios is observed throughout the burnup cycle. The lowest levels for these ratios are reached at 60.00 MWd/kgU, with ratios of 8.29 wt% and 9.42 wt% respectively.

3.2. Fuel cycle analysis

This section examines reactor operation with ^{241}Am-doped nuclear proliferation-resistant fuel cycle and IFBA-equipped standard reactor operation. In Table 3, data relating to the SIMULATE3 fuel cycle analysis for the scenario IFBA used standard reactor operation, including the beginning of the cycle's (BOC) critical boron concentration (CBC), the CBC at 10 MWd/kgU, BOC-k_{eff}, nuclear enthalpy rise hot channel factor ($F_{\Delta H}$) and heat flux hot channel factor (F_Q) throughout the cycle, cycle Burnup, and cycle EFPDs, are presented. In contrast, in Table 3, data for the scenario where ^{241}Am-doped fuel is utilised are displayed.

When Table 3 is examined, it is observed that the scenario using IFBA-equipped fuel reaches approximately 20.93 MWd/kgU burnup and 530.5 EFPDs in the first cycle. Due to the fuel loading pattern employed in subsequent cycles, some fuels with 3.60 wt% and 4.65 wt% uranium enrichment needed to be discharged, leading to a drop in burnup to approximately 18.24 MWd/kgU and 462.3 EFPDs in the second cycle. It should be noted that to address this situation and achieve equivalent burnup and EFPDs across all cycles, a transitioning fuel loading pattern needs to be utilised. In the following cycles, it is noticed that the cycle burnup and EFPDs increased, stabilising by the sixth cycle. In the sixth cycle, a burnup of 20.363 MWd/kgU and 516.3 EFPDs is observed.

Upon the examination of Table 4, it is observed that in the first cycle of the scenario where ^{241}Am-doped fuel is used, a burnup of 19.32 MWd/kgU is reached, amounting to 489.8 EFPDs. Likewise, due to the fuel loading pattern in the second cycle, a sharp drop in EFPDs is noted, but equilibrium is achieved over 6 cycles. In the sixth cycle, a burnup of 19.33 MWd/kgU is achieved, resulting in 490.0 EFPDs.

Comparing Table 3 and Table 4, it is seen that in the scenario of using IFBA-free ^{241}Am-doped fuel, the EFPD is approximately 7.6 % (40.7 EFPDs) lower than that of the IFBA-equipped fuel. On the other hand, when the sixth cycles of both scenarios are compared, it is observed that the difference between the EFPDs drops to 26.3 EFPDs. Of course, the reason for these declines is due to the need for a high amount of neutron-absorbing ^{241}Am to achieve a ratio of over 6.00 wt% of ^{238}Pu/TotPu in the fuel composition throughout the burnup. The particularly higher decline observed in the first cycle is because a higher proportion of ^{241}Am was used in the first cycle compared to other cycles.

When the CBC values of both scenarios are examined, it is observed that the BOC CBC in the first cycle of the scenario using IFBA-equipped fuel is approximately 1,515 ppm. Meanwhile, in the scenario using ^{241}Am-doped fuel, the BOC CBC of the first cycle is found to be approximately 835 ppm. This value is consistent with the reduction seen in the first cycle EFPDs when ^{241}Am-doped fuel is used. In comparison, when the BOC CBCs of the sixth cycle of both scenarios are compared, an

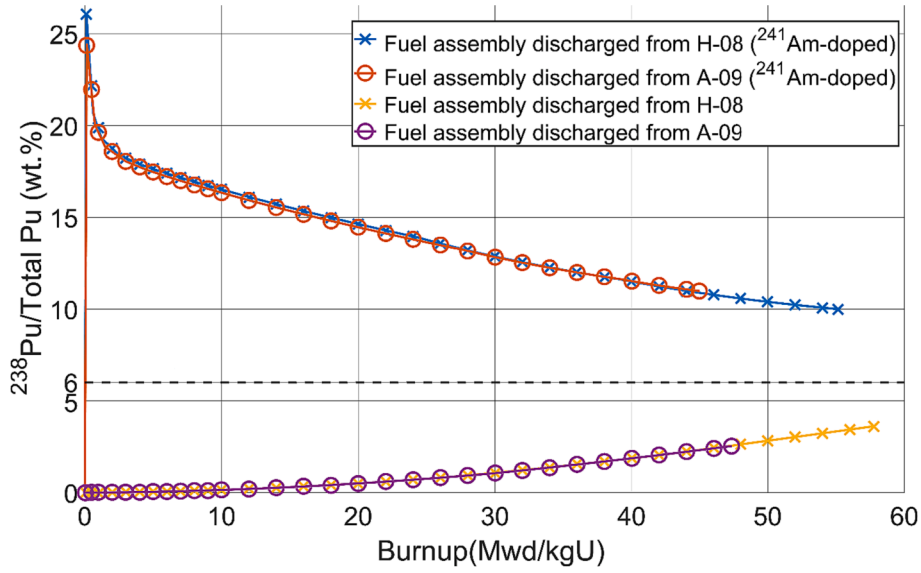

Fig. 5. ^{238}Pu/TotPu ratio of the fuel assemblies discharged at the end of cycle 6 for both IFBA-equipped and ^{214}Am-free scenarios.

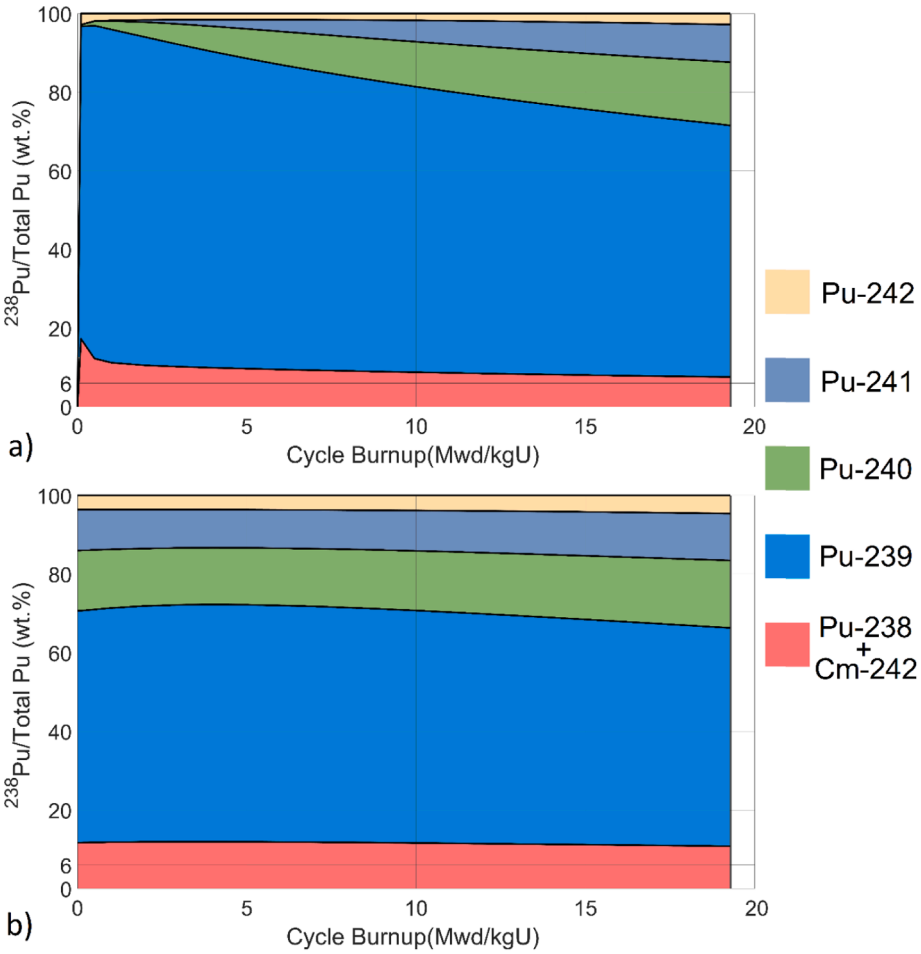

Fig. 6. Mass fraction of different Pu isotopes as a function of burnup.

increase of about 11 % (217 ppm) to approximately 1,961 ppm is noted with the use of ^{241}Am-doped fuel.

On the other hand, when $F_{\Delta H}$ values are compared, a decrease of 1.3 % in the first cycle and an increase of 0.9 % in the sixth cycle are observed. Additionally, when F_Q values are compared, increases of approximately 5 % are seen in both the first and sixth cycles. However, it should be noted that both $F_{\Delta H}$ and F_Q values remain within the design and operation limits for both scenarios.

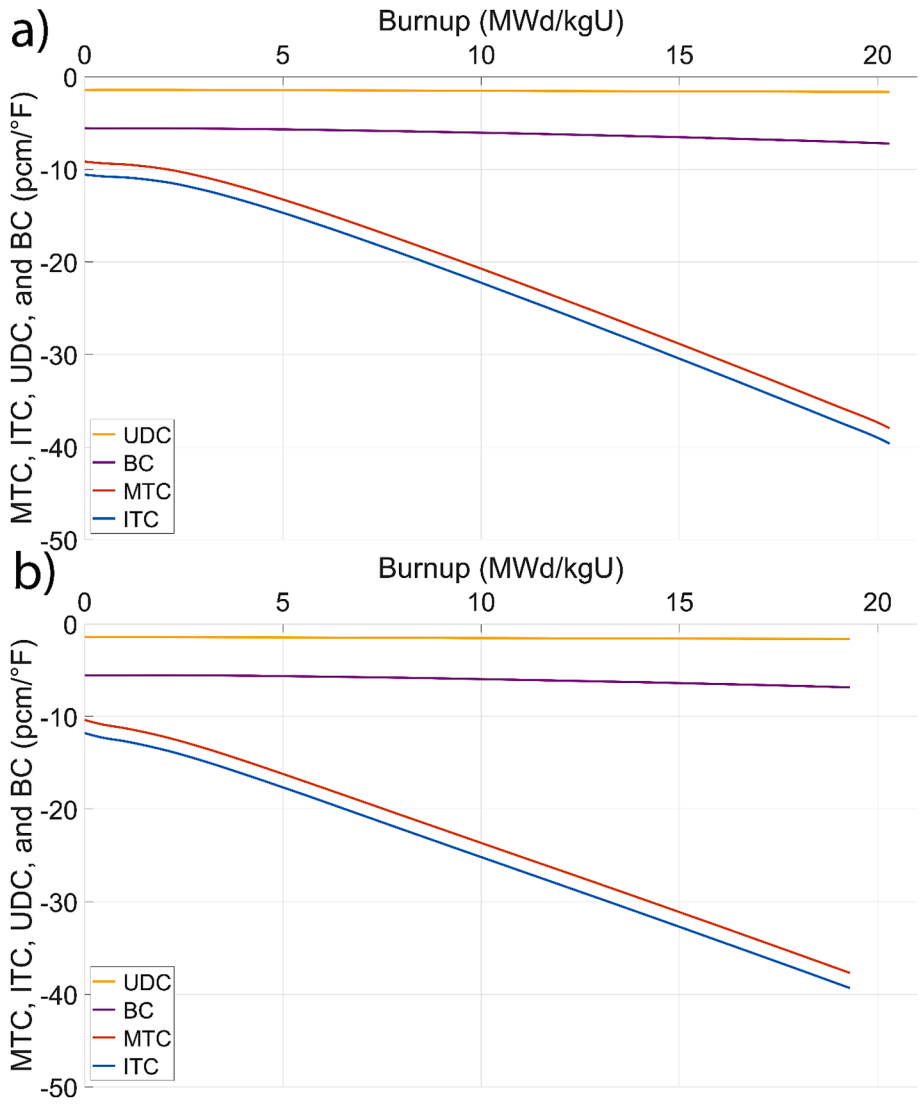

Fig. 7. Comparison of reactivity feedback parameters - UDC, BC, MTC, and ITC - for the 6th cycles: (a) IFBA-equipped case and (b) ^{241}Am-doped case.

Table 5
MTC, ITC, UDC and BC values of 6th cycles both for BOC and EOC.

		MTC (pcm/°F)	ITC (pcm/°F)	UDC (pcm/°F)	BC (pcm/°F)
Core with IFBA-equipped fuel	BOC	−9.17	−10.58	−1.43	−5.56
	EOC	−37.96	−39.62	−1.64	−7.22
Core with ^{241}Am-doped fuel	BOC	−10.37	−11.79	−1.43	−5.55
	EOC	−37.73	−39.37	−1.63	−6.86

Table 6
BOC and EOC shutdown margins.

	Cycle	BOC (pcm)	EOC (pcm)
Core with IFBA	Cycle 01	−4,329	−2,316
	Cycle 06	−3,706	−2,351
Core with ^{241}Am-doped fuel	Cycle 01	−3,902	−2,325
	Cycle 06	−3,466	−2,311

3.3. Pu breeding behaviour

For ensuring control overachieving a proliferation-resistant fuel cycle, the ^{238}Pu/TotPu ratios of the scenarios in which ^{241}Am-doped fuel is used and IFBA-equipped fuel is employed were monitored throughout all cycles.

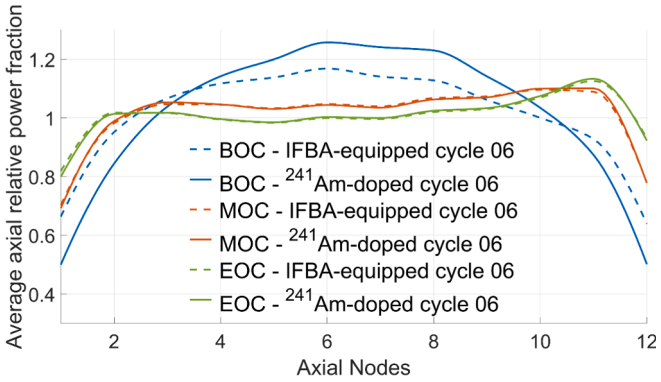

Fig. 8. Average axial relative power distribution curves (top to bottom) in the 6th cycle.

In Fig. 4, the average ^{238}Pu/TotPu ratio within the reactor during the 1st, 2nd, and 6th cycles for the scenario where ^{241}Am-doped fuel is used and for the scenario where IFBA-equipped fuel is employed is displayed. As can be seen in Fig. 4, in the scenario where IFBA-equipped fuel is

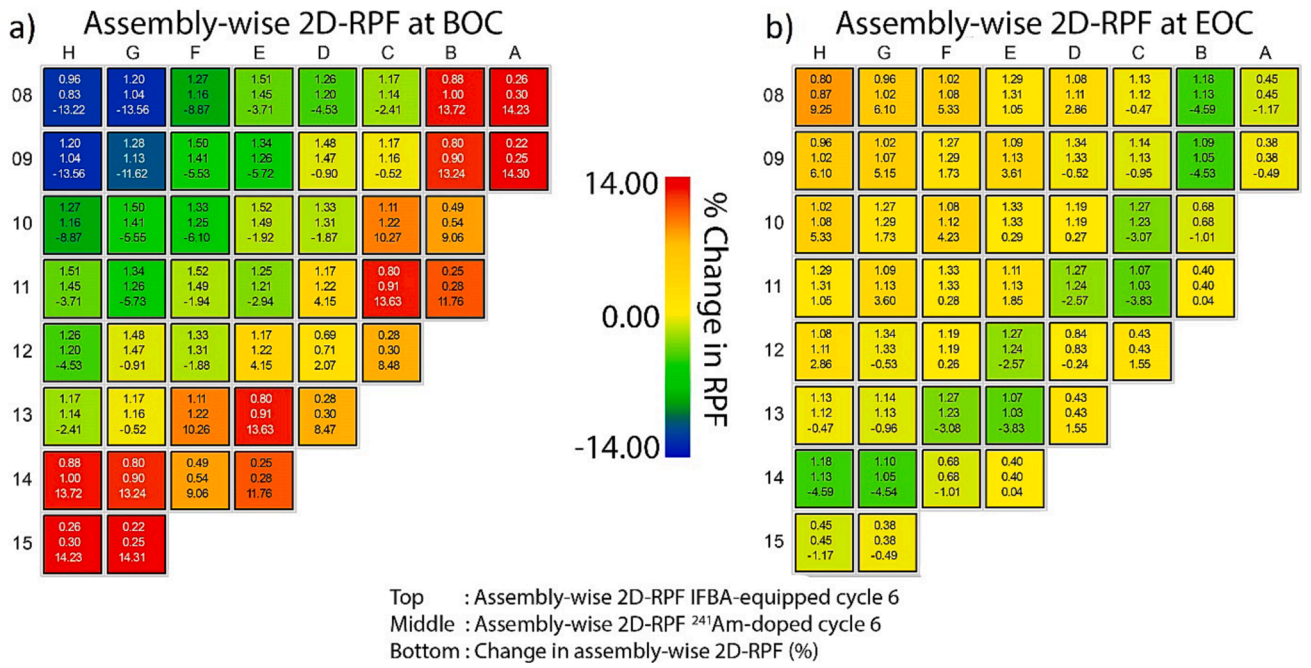

Fig. 9. Comparison of relative power fractions on a 2D assembly basis for the 6th cycle, contrasting IFBA-equipped case with ^{241}Am-doped case: (a) at BOC and (b) at EOC.

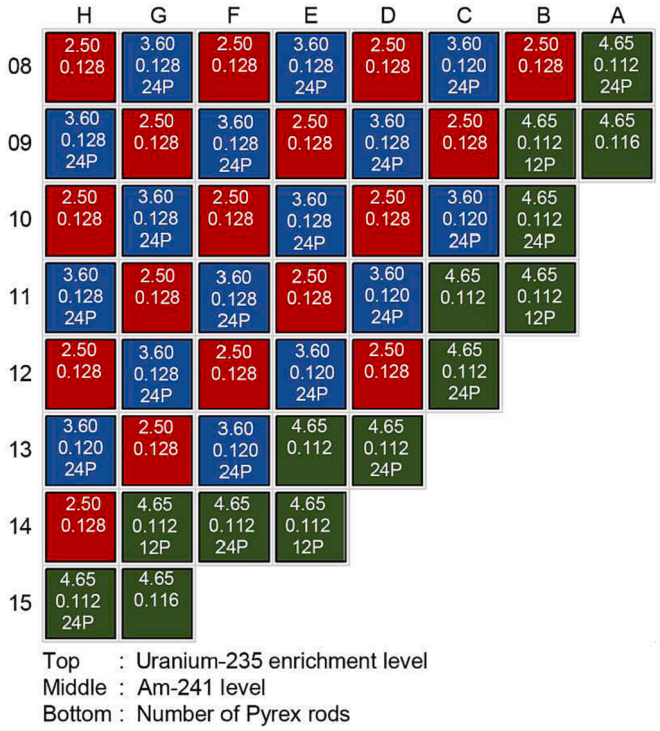

Fig. A1. Cycle 1 fuel loading pattern, enrichment levels, ^{241}Am levels (for the ^{241}Am-doped fuel), and number of Pyrex rods.

used, the ^{238}Pu/TotPu ratio gradually increases in the first cycle, reaching only 0.69 wt% by the end of the cycle. This ratio then rises to 1.48 wt% at the end of the second cycle and to 2.04 wt% at the end of the sixth cycle.

On the other hand, when the curves corresponding to the scenario in which ^{241}Am-doped fuel is used are examined, it is seen that the average ^{238}Pu/TotPu ratio inside the reactor peaks at 0.10 MWd/kgU, reaching 17.29 wt% in the first cycle. Subsequently, this ratio drops to 12.29 wt% at 1.00 MWd/kgU and continues to decline gradually for the rest of the cycle, ending at 7.57 wt%. Meanwhile, at the beginning of the second cycle, the ^{238}Pu/TotPu ratio stands at 7.48 wt%. it should be noted that the discrepancy between the ^{238}Pu/TotPu ratio at the end of the first cycle and the beginning of the second cycle is attributed to the fact that these curves depict the average ^{238}Pu/TotPu ratio within the reactor, and the fresh fuels do not possess a Pu content at the beginning of the second cycle.

Following this, the ^{238}Pu/TotPu ratio increases with the progression of burnup, reaching its peak value within the cycle at 10 MWd/kgU (9.13 wt%) in the second cycle. It then slightly decreases, settling at 8.88 wt% by the end of the cycle. Furthermore, at the beginning of the sixth cycle, the ^{238}Pu/TotPu ratio stands at 11.68 wt%. Although this ratio ascends to its highest level within the cycle at 3.00 MWd/kgU, achieving 11.95 wt%, then it subsequently reduces to 10.81 wt% at the end of the cycle. In summary, by using ^{241}Am-doped fuel, a ^{238}Pu/TotPu ratio of over 6.00 wt% can be achieved in all cycles, enabling the attainment of a proliferation-resistant fuel cycle.

To ensure the achievement of a proliferation-resistant fuel cycle, all fuel assemblies used in fuel cycles were individually monitored to ascertain that they maintained a ^{238}Pu/TotPu ratio of over 6.00 wt%. As a case in point, the ^{238}Pu/TotPu ratios throughout the lifetimes of the fuel assemblies with the highest and lowest burnups, loaded into the reactor during the 4th fuel cycle and discharged at the end of the 6th fuel cycle, are presented in Fig. 5 for both the scenario where ^{241}Am-doped fuel is used and the scenario where IFBA-equipped fuel is employed.

Upon examining Fig. 5, in the scenario where ^{241}Am-doped fuel is used, the ^{238}Pu/TotPu ratios of the fuels discharged from the H-08 and A-09 locations inside the reactor (see Fig. 1) are 26.08 wt% and 24.42 wt % at the beginnings of their lifetimes, respectively. Following this peak, there is a rapid decline in the ^{238}Pu/TotPu ratio, succeeded by a gradual decrease. Subsequently, the fuel discharged from the H-08 location diminishes to 9.99 wt% by the end of its lifetime (corresponding to 55.14 MWd/kgU), while the fuel assembly discharged from A-09 possesses a ^{238}Pu/TotPu ratio of 10.98 wt% at the end of its lifetime (corresponding to 44.97 MWd/kgU).

In Fig. 6, a graph displaying the ratio of various Pu isotope to the

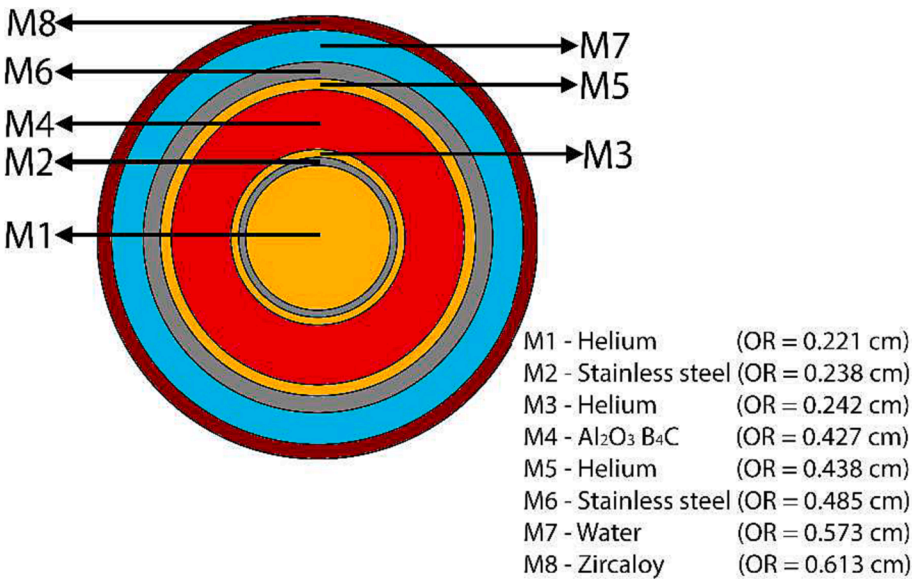

Fig. A2. Cross-section of Pyrex burnable poison rod.

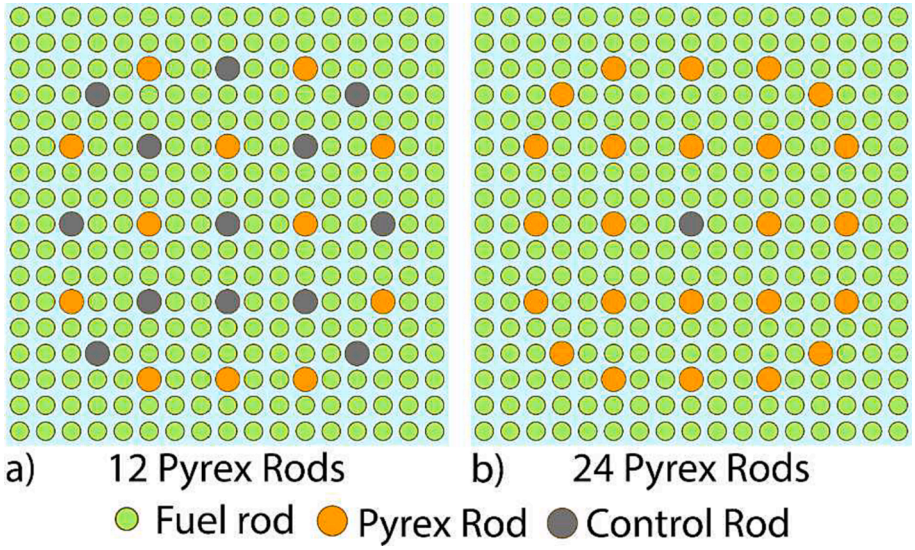

Fig. A3. Distrubution of Pyrex rods.

total Pu amount in the scenario where ^{241}Am-doped fuel is used is provided. Specifically, Fig. 6a depicts the curves corresponding to the 1st cycle, while Fig. 6b illustrates those related to the 6th cycle.

In Fig. 6a, the ratio of ^{239}Pu/TotPu is initially found to be at 79.46 wt %. Later, it peaks at 84.71 wt% at 1.0 MWd/kgU. By the end of the cycle, this ratio has been reduced to 63.97 wt%. In contrast, the ratios of ^{240}Pu/TotPu and ^{241}Pu/TotPu are noted to consistently increase throughout the cycle, concluding at values of 16.10 wt% and 9.58 wt% respectively. The ^{242}Pu/TotPu ratio is revealed to surge to 2.81 wt% at the beginning of the cycle, then drop to 1.52 wt% at 4.0 MWd/kgU, and finally rise to approximately 2.79 wt% by the end of the cycle.

Upon examination of Fig. 6b, the ratio of ^{239}Pu/TotPu is seen to peak at 60.31 wt% at 4.0 MWd/kgU. At this burnup point, the lowest recorded values for the ratios of ^{240}Pu/TotPu, ^{241}Pu/TotPu, and ^{242}Pu/TotPu are 15.51 wt%, 9.75 wt%, and 3.66 wt% respectively. On the other hand, by the end of the cycle, the most diminished ratio of ^{239}Pu/TotPu is identified as 55.478 wt%. The highest values for ^{240}Pu/TotPu, ^{241}Pu/TotPu, and ^{242}Pu/TotPu are observed at the cycle's termination, registering at 17.16 wt%, 11.94 wt%, and 4.61 wt% respectively. As such, during any stage of the reactor's function, it is concluded that the fuel is not amenable to re-purposing with weaponizing aims.

3.4. Reactivity feedback parameters, shutdown margin and power profiles

Reactivity feedback parameters, which hold significance in the safe operation of the reactor, such as BC, UDC, ITC, MTC were examined. In Fig. 7, curves related to reactive feedback parameters are presented for the sixth cycle of scenarios where fuel equipped with IFBA is used (Fig. 7a) and fuel without ^{241}Am is used (Fig. 7b). Moreover, the BOC and the end of the cycle (EOC) values of these parameters are displayed in Table 5.

For the cycle where fuel equipped with IFBA is used, it is observed that the initial values of MTC and ITC are −9.17 pcm/°F and −10.58 pcm/°F, respectively. A gradual decline in these parameters is observed throughout the cycle, and by its end, they are found to be −37.73 pcm/°F and −39.37 pcm/°F, respectively. When the uniform Doppler coefficient and boron coefficient are analysed, they are found to be −1.43 pcm/°F and −5.56 pcm/°F at the beginning of the cycles, respectively. A

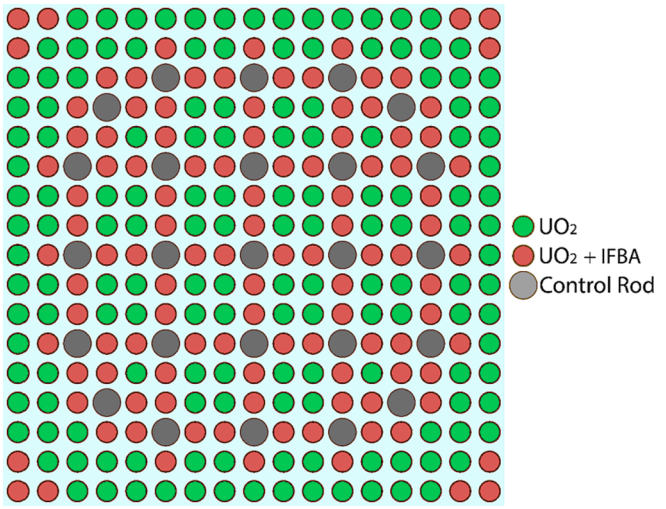

Fig. A4. Distrubution of IFBA rods (Ames Ii et al., 2010).

consistent reduction in these parameters is noticed over the cycle's duration, culminating in values of −1.64 pcm/°F and −7.72 pcm/°F by the cycle's end, respectively.

On the other hand, when the graph relating to the cycle with ^{241}Am-doped fuel is inspected, it is noted that both MTC and ITC commence the cycle at values approximately 1 pcm/°F more negative than in the scenario with fuel equipped with IFBA. When the values at the end of the cycle are reviewed, no significant change is observed.

In the scenario where ^{241}Am-doped fuel is utilised, it is noted that no significant alterations are seen in UDC and BC in comparison to the fuel scenario with IFBA-equipped fuel. Specifically, at the beginning of the cycle, the parameters are identified to be −1.43 pcm/°F and −5.56 pcm/°F, respectively. Over the cycle, relatively small reductions in these values are observed, finalising at −1.64 pcm/°F and −7.22 pcm/°F, respectively, by the cycle's end. In summation, no adverse impact that on the reactivity feedback parameters that could compromise the reactor's safe operation is detected.

Within the framework of guaranteeing the nuclear reactor's safe and consistent operation, the significance of the shutdown margin is underscored as an essential feature aiding in proficient system management and regulation. For the assessment of the influence on the shutdown margin when ^{241}Am-doped fuel is used, computations of the shutdown margins for scenarios that use both IFBA-equipped fuel and ^{241}Am-doped fuel were carried out. Table 6 displays the shutdown margin values related to the first and sixth cycles for these scenarios.

In both situations, elevated shutdown margins in the first cycles compared to the sixth cycles are noted, which is anticipated due to the use of Pyrex rods within the system. Specifically, in the scenarios where IFBA-equipped fuel is utilised, the shutdown margin in the first cycle is found to be roughly 14.39 % (623 pcm) greater than in the sixth cycle. In a similar manner, when ^{241}Am-doped fuel is used, the shutdown margin for the first cycle is roughly 11.17 % (436 pcm) greater than for the sixth cycle. Additionally, insignificant changes in the end-of-cycle shutdown margins are detected in both scenarios.

Conversely, when comparing scenarios that utilise ^{241}Am-doped fuel with that use IFBA-equipped fuel, it is determined that the scenario using ^{241}Am-doped fuel exhibits lower shutdown margin values at the beginning of the first and sixth cycles. For instance, in the first cycle of the two scenarios, a difference of 9.86 % (427 pcm) is observed, while in the sixth cycle of those scenarios, there is a discrepancy of 6.48 % (240 pcm). Taking into account the safety limit of the modelled reactor, these differences are not of significant importance. Furthermore, when the EOC shutdown margins of both scenarios are compared, no noteworthy variation is detected.

In Fig. 8, 2D average axial relative power fraction curves at the beginning, middle, and end of the 6th cycle are displayed for scenarios using both ^{241}Am-doped fuel and IFBA-equipped fuel. Upon examining Fig. 8, it becomes evident that in the scenario using IFBA-equipped fuel, the BOC 2D average axial relative power fraction has a flatter distribution. However, in the scenario with ^{241}Am-doped fuel, a discernible discrepancy in power distribution between the middle section of the reactor and the top and bottom parts is observed. Of course, this is attributed to the preference for 15.24 cm BA-free zones in the top and bottom sections of the fuel rods in the IFBA-equipped fuel scenario to achieve a flatter profile. In the ^{241}Am-doped fuel, all the fuel used did not employ a ^{241}Am-free zone so that a ^{238}Pu/TotPu ratio exceeding 6.00 wt% could be achieved. Yet, for a smoother power profile, the ^{241}Am concentration can be reduced in the top and bottom parts of the fuel rods while increasing it in the middle section. As shown in the previous section, since the ^{238}Pu/TotPu ratio within the fuel assemblies is relatively above 6.00 wt%, reducing the ^{241}Am concentration for this purpose will still yield values above the desired ^{238}Pu/TotPu ratio. Additionally, when examining the 2D average axial relative power fraction curves for both scenarios at mid-cycle and end of the cycle, it is observed that no significant changes have occurred.

In Fig. 9, values for the assembly-wise 2D relative power fraction at the beginning of the cycle (Fig. 9a) and end of the cycle (Fig. 9b) for scenarios utilising IFBA-equipped and ^{241}Am-doped fuel in their 6th cycle are presented.

Using ^{241}Am-doped fuel, changes in the assembly-wise 2D relative power fraction were observed at the BOC. It was found that the most significant decrease occurred in the once-burned fuels at G-08 and H-09 locations, bringing the PPF closer to 1. While a decrease was also noted in the twice-burned fuel at the H-09 location, this change moved the RPF away from 1. On the other hand, the most significant increase was observed in the twice-burned fuels at the A-09 and G-15 locations, moving the RPF closer to 1. Overall, it was noted that while the majority of these changes tended to bring the RPF closer to the ideal value of 1, such changes were deemed negligible in terms of reactor operation.

On the other hand, when changes in the EOC assembly-wise 2D relative power fraction resulting from the use of ^{241}Am were examined, it was observed that the most noticeable increase took place in the twice-burned fuel at the H-08 location, moving the RPF closer to 1. Additionally, the most notable decrease was found in the fresh fuels located at the B-08 and H-14, also tending to bring the RPF nearer to 1. Furthermore, changes occurring in the EOC 2D relative power fraction due to the use of ^{241}Am generally resulted in PRF values closer to 1.

4. Conclusions

In the study, the possibility of using nuclear proliferation-resistant fuel in a PWR without the need for an additional burnable absorber was explored by incorporating ^{241}Am into the fuel composition. It was conclusively shown that such a proliferation-resistant fuel cycle could be established within design and safety limits without the inclusion of extra BA.

In the investigation, scenarios using IFBA-equipped fuel were compared with those using ^{241}Am-free fuel. Consideration was given to factors like the heat flux hot channel factor and the nuclear enthalpy rise hot channel factor. Reactivity feedback parameters, including the moderator temperature coefficient, isothermal temperature coefficient, critical boron concentration, boron coefficient, and uniform Doppler coefficient, were also examined. Furthermore, the impact of ^{241}Am-doped fuel on the power profile and shutdown margin was analysed.

It was found that the utilization of ^{241}Am-doped fuel does not adversely affect the heat flux hot channel factor or the nuclear enthalpy rise hot channel factor during the operation of the reactor. Nevertheless, increases within the design and operational limits were observed.

When ^{241}Am-doped fuel is utilized, negative values of 1 pcm/°F for both the moderator temperature coefficient and the isothermal temperature coefficient are observed at the start of the cycle; however, these

can be considered negligible. Furthermore, no significant alterations were noted in the uniform Doppler coefficient and the boron coefficient parameters. As a result, it was determined that the use of ^{241}Am-doped fuel did not result in any changes that would compromise the safety of reactor operation in terms of reactivity feedback parameters.

While the impact of ^{241}Am on the assembly-wise 2D relative power fraction is relatively minor, in fuel assemblies with the most pronounced changes, there's typically a positive influence that tends to align the RPF more closely with its optimal value.

Regarding the shutdown margin, which is one of the most crucial parameters for ensuring safe and reliable reactor operation, a decrease at the BOC was noted with the use of ^{241}Am-doped fuel. Nonetheless, this change did not present a threat to the shutdown margin, as the computed value for the shutdown margin consistently stayed well above the minimum criteria.

In conclusion, it was shown that, depending on variations related to factors like fuel composition, loading pattern, and batch number, incorporating ^{241}Am into the fuel composition could lead to nuclear proliferation-resistant fuel. This benefit, however, comes with a trade-off of approximately 5 % EFPDs, which equates to roughly 26 EFPDs.

Given the innovative nature of the fuel design, comprehensive research should be carried out to determine the potential changes in thermal behaviour caused by the utilization of ^{241}Am-doped fuel. Secondly, attention should be directed towards optimizing reactor core configurations to possibly lengthen EFPDs and enhance burnup rates. Lastly, economic analyses of scenarios using IFBA-equipped fuel versus ^{241}Am-doped fuel should be conducted. It should be investigated how much of the financial loss resulting from the reduction in EFPDs can be compensated by the financial gains achieved by not using IFBAs.

5. Data availability

The produced data supporting the findings of this paper are available from the corresponding author upon reasonable request.

Conflicts of interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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CRediT authorship contribution statement

Mustafa J. Bolukbasi: Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Conceptualization. **Marat Margulis:** Writing – original draft, Supervision, Resources, Funding acquisition, Formal analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendices A.

In Fig. A1, the fuel loading pattern utilised in the first cycle of the scenarios with IFBA-equipped fuel and 241Am-doped fuel is depicted. In this figure, the enrichment levels of uranium in the fuel assemblies are indicated. The percentage (in wt.%) of 241Am in the fuel composition for the 241Am-doped fuel model is also shown. Moreover, the number of Pyrex rods contained in the fuel assemblies is illustrated.

Fig. A2 illustrates the cross-sectional view of the Pyrex rods utilized in the initial cycles, whereas Fig. A3 depicts how these Pyrex rods are arranged within the fuel assemblies.

Fig. A4 shows the distribution of IFBAs (in 112 rods) used in fresh fuels within the fuel assembly

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