Hindawi
Science and Technology of Nuclear Installations
Volume 2019, Article ID 5769147, 12 pages
https://doi.org/10.1155/2019/5769147

Research Article

Study on Transmutation of Minor Actinides as Burnable Poison in VVER-1000 Fuel Assembly

Vinh Thanh Tran,[1] **Hoai-Nam Tran**,[2] **Huu Tiep Nguyen**,[1] **Van-Khanh Hoang**,[1] **and Pham Nhu Viet Ha**[1]

[1]*Institute for Nuclear Science and Technology, VINATOM, 179 Hoang Quoc Viet Str., Cau Giay Dist., Hanoi 100000, Vietnam*
[2]*Institute of Fundamental and Applied Sciences, Duy Tan University, Ho Chi Minh City 700000, Vietnam*

Correspondence should be addressed to Vinh Thanh Tran; tvthanh@vinatom.gov.vn and Pham Nhu Viet Ha; hapnv0915@gmail.com

Received 29 March 2019; Revised 8 August 2019; Accepted 21 August 2019; Published 3 November 2019

Guest Editor: Dimitrios Tsaoulidis

Thermal reactors have been considered as interim solution for transmutation of minor actinides recycled from spent nuclear fuel. Various studies have been performed in recent decades to realize this possibility. This paper presents the neutronic feasibility study on transmutation of minor actinides as burnable poison in the VVER-1000 LEU (low enriched uranium) fuel assembly. The VVER-1000 LEU fuel assembly was modeled using the SRAC code system, and the SRAC calculation model was verified against the MCNP6 calculations and the available published benchmark data. Two models of minor actinide loading in the LEU fuel assembly have been investigated: homogeneous mixing in the UGD (Uranium-Gadolinium) pins and coating a thin layer to the UGD pins. The consequent negative reactivity insertion by minor actinides was compensated by reducing the gadolinium content and boron concentration. The reactivity of the LEU assembly versus burnup and the transmutation of minor actinide nuclides were examined in comparison with the reference case. The results demonstrate that transmutation of minor actinides as burnable poison in the VVER-1000 reactor is feasible as minor actinides could partially replace the functions of gadolinium and boric acid for excess reactivity control.

1. Introduction

It is recognized that the negative fames of nuclear energy, i.e., the current nuclear power plants (NPPs) being operated worldwide for electricity generation, are the release of radioactive materials under normal, abnormal, or accident conditions and the by-products of highly radioactive, long-lived spent nuclear fuels. Hence, a closed nuclear fuel cycle has been considered as the best option to overcome the issues with disposal of used fuel to geologic repository. On average, a light water reactor (LWR) with electric capacity of 1000 MWe produces 20–30 metric tons of spent nuclear fuel annually, which consist of approximately 95 wt.% uranium, 1 wt.% plutonium, 4 wt.% fission products, and minor actinides (MAs) [1, 2]. In the used fuel, the trans-uranic elements, i.e., plutonium and MAs, dominate the decay heat load to the repository and cumulative long-term radiotoxicity to the environment. To lessen the burden for disposal and storage of spent nuclear fuel and to reduce its cumulative radiotoxicity to the environment, separation and transmutation of the plutonium and MAs in the used fuel are indispensable [3]. It has been realized that the transmutation of these actinides into either short-lived fission products or valued fissile or stable isotopes can be accomplished in fast reactors, subcritical reactors, or thermal reactors [1, 2, 4–8].

Fast reactors and subcritical reactors have been studied as the most potential candidates for transmutation of the actinides, thanks to their hard neutron spectra. However, these future technologies are still not mature nowadays and require at least several decades or even longer to be well proved and deployed on a large commercial scale. This

indicates that an interim solution to the used nuclear fuel, i.e., transmutation of the plutonium and MAs, is needed in near-term until the commercial deployment of fast reactors or subcritical reactors in future. LWRs, e.g., pressurized water reactors (PWRs), which are the most proven nuclear energy technologies, have been thus extensively studied as well for their transuranic transmutation capability with consideration to their numerous numbers currently being operated commercially in the world. Namely, once the actinide transmutation capability of LWRs is demonstrated, the current LWRs can be immediately deployed on a large scale to destroy the actinides for the sake of generating power and reducing the burden from disposal and storage of used fuel. In this regard, the MA transmutation in LWRs has drawn much attention so far to serve as a near-term solution to the issues with spent fuel and various methods of loading MAs into the LWRs have been investigated to realize such possibility. Nonetheless, most of the studies are reported with the Western PWRs [9–16]. Similar studies with the Russian water-water energy reactors (VVERs) have rarely been found in the literature although a large number of NPPs based on the VVER technologies are being operated in various East European and Asian countries [17, 18].

The VVER reactor is obviously a potential candidate for transmutation of actinides in the spent fuel stock-pile, and various methods of loading and burning transuranic elements in the Western PWRs may be adopted similarly to the Russian VVERs. In the past studies, transmuting MAs in the burnable poison rods [19, 20] or in some other locations in the PWR fuel assemblies has been found technically feasible and recommended as potential transmutation methods for LWRs, especially the unique advantage of loading MAs to partially replace the excess reactivity control functions of gadolinium and boric acid.

The present study therefore aims at investigating the neutronic feasibility of MA transmutation in a VVER-1000 low enriched uranium (LEU) fuel assembly [21]. The goal is to determine how efficient the MAs (neptunium, americium, and curium) recycled from spent fuel can be transmuted in the VVER-1000 fuel assembly. The MA loading into the VVER-1000 fuel assembly will be performed without significant modification of the assembly configuration, because any significant change of the fuel assembly design will lead to a penalty in the cost for fuel fabrication process and respective changes in reactor core design. The SRAC code system [22] is used for modeling the VVER-1000 LEU fuel assembly based on the ENDF/B-VII.0 library. The calculation model with SRAC is verified against the Monte Carlo calculations with the MCNP6 code [23] and the available published benchmark results. In recent publications, the burnable absorber rods have been suggested as potential locations for loading and burning MAs [18–20] and therefore two approaches are examined in this study: (a) MAs are mixed homogeneously in the UGD (Uranium-Gadolinium) pellets and (b) a coating layer of MAs is included to the UGD pellets. The constraint for these MA loadings is to ensure insignificant change in the reactivity of the fuel assembly while providing considerable MA transmutation rate.

The paper is organized as follows. The analysis model and methods of loading MAs into the VVER-1000 LEU fuel assembly are given in Section 2. The verification of the SRAC model for the VVER-1000 LEU fuel assembly is presented in Section 3. The results and feasibility of burning MAs in the VVER-1000 LEU fuel assembly are shown and discussed in Section 4. Lastly, concluding remarks and further works are represented in Section 5.

2. Calculation Methodology

The VVER-1000 LEU fuel assembly specified in the OECD VVER-1000 LEU and MOX (mixed oxide) Assembly Computational Benchmark [21] is utilized in the present investigation to examine the feasibility of MA transmutation as burnable poison in the VVER-1000 reactor. The configuration and main design parameters of the VVER-1000 LEU fuel assembly are shown in Figure 1 and Table 1, respectively. It is recalled that the benchmark model is designed to verify the computational codes for VVER-1000 LEU and MOX fuel calculations in support for the weapon grade plutonium disposition mission. It consists of two different hexagonal fuel assemblies: a uniform LEU fuel assembly and a profiled MOX fuel assembly. The LEU assembly consists of 300 fuel pin cells with 3.7 wt.% ^{235}U, 12 UGD pin cells with 3.6 wt.% ^{235}U, and 4 wt.% Gd_2O_3, 18 water filled guide tubes for control insertion, and one central water filled instrumentation tube. The MOX assembly consists of 138 fuel pin cells with 4.2 wt.% fissile Pu in the central region, surrounded by 96 fuel pin cells with 3 wt.% fissile Pu and 66 fuel pin cells with 2 wt.% fissile Pu at the outermost region, 12 UGD pin cells with 3.6 wt.% ^{235}U and 4 wt.% Gd_2O_3, 18 water filled guide tubes, and one central water filled instrumentation tube. These two assemblies are representative of the advanced designs under active R&D in Russia for VVER-1000 reactors and similar to the designs that are expected to be used in the plutonium disposition mission. In these assembly designs, the burnable absorber Gd_2O_3 is mixed with UO_2 to form the UGD pin cells for excess reactivity control at the beginning of cycle.

The VVER-1000 LEU fuel assembly is modeled in this work using the SRAC code system. In the SRAC simulation, the one-sixth of the LEU fuel assembly is modeled with the PIJ module (see Figure 2), the fuel burnup calculations are performed with the BURN-UP module, and the 107 energy groups based on the ENDF/B-VII.0 nuclear data library are used. The LEU fuel assembly modeled with SRAC is verified against the Monte Carlo code MCNP6 calculations and the published benchmark data for the S1 state, i.e., the normal operating poisoned state with ^{135}Xe and ^{149}Sm equilibrium concentrations. The parameters to be compared include the infinite multiplication factor (k-inf) of the fuel assembly versus burnup and nuclide concentrations.

According to [24], a combination of VVER-1000 and fast reactors was recommended for transmuting actinides recycled from spent fuel. The VVER-1000 reactor can be used for burning the plutonium in the form of MOX fuel. The fast reactor can be used for burning the MAs recycled from spent fuels of the VVER-1000 and fast reactors and the

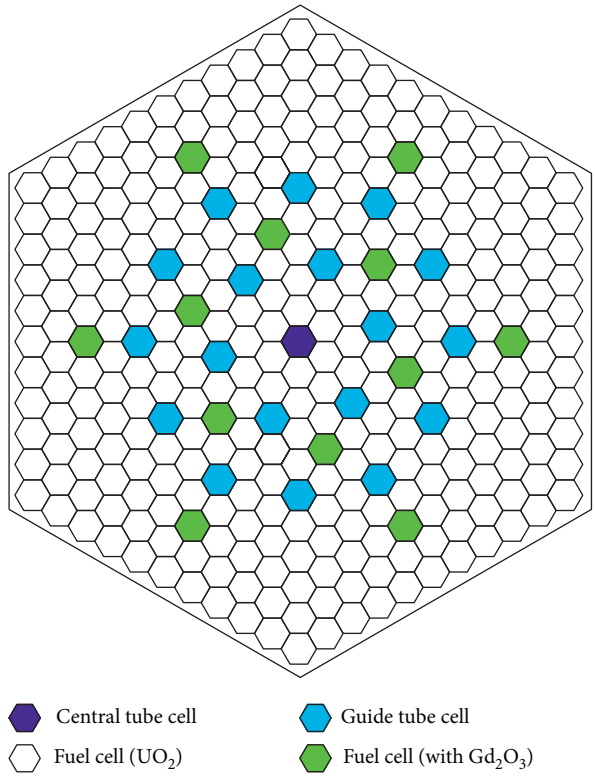

Figure 1: Configuration of the VVER-1000 LEU fuel assembly.

Table 1: Main design parameters of the VVER-1000 LEU fuel assembly.

Parameter	Value
Number of UO_2 fuel cells	300
Number of fuel cells with Gd	12
Number of guide tubes	18
Number of central tubes	1
Fuel cell inner radius (cm)	0.3860
Fuel cell outer radius (cm)	0.4582
Central tube cell inner radius (cm)	0.5450
Central tube cell outer radius (cm)	0.6323
Pin pitch (cm)	1.2750
Fuel assembly pitch (cm)	23.6
Fuel temperature (K)	1027.0
Nonfuel temperature (K)	575.0
^{235}U enrichment (wt.%)	3.7
Gd_2O_3 density (g/cm^3)	7.4

plutonium recycled from the spent fuel of the fast reactor. Another study showed the possibility of actinide transmutation in the VVER-440 reactor by loading the non-uranium pins consisting of actinides recycled from spent fuel to the periphery of the VVER-440 fuel assemblies [17]. The replacement of UGD pins in the VVER-1000 reactor by target elements containing a mixture of graphite and transmuted actinides has been also studied for the transmutation of americium and curium, and it was reported that the VVER-1000 reactor can operate in the self-service mode, i.e., the mode of transmutation when the number of transmuted nuclides is equal to the amount of their buildup in the fuel of the reactor itself [18]. It indicates that the VVER reactors can be used not only for the weapon grade plutonium disposition mission but also for the burning of actinides recycled from used fuel. Thus, the MA transmutation capability of VVER reactors should be carefully examined.

In this investigation, we intend to load the MAs in the UGD pins of the VVER-1000 LEU fuel assembly for their transmutation without significant change in the fuel assembly configuration. The purpose is to investigate the transmutation capability of the VVER-1000 LEU fuel assembly. To this end, we consider two approaches to load the MAs into the fuel assembly while tuning the gadolinium content and boron concentration: (1) mixing MAs homogeneously with UO_2 and Gd_2O_3 in the UGD pellets and (2) coating a thin layer of MAs around the UGD pellets. The rationale is that MAs can partially act as burnable poison and thus can partially replace the functions of the gadolinium and boric acid to control excess reactivity of the fuel assembly [19, 20].

It is recalled that the greatest industrial experience exists for UO_2 fuels for LWRs without any special shielding consideration in the fabrication process. Additionally, MOX fuels, which are fabricated in glove boxes with steel or lead shielding, have been mastered at the industrial level, especially with highly automated plants operating in France and the UK [1, 2]. However, the fabrication of MA bearing fuels with high gamma and neutron doses from MAs requires extra biological protection in the form of lead (for gamma radiation) and a combination of water, lead, and cadmium or boron (for neutron radiation). Also, adequate shielding should be provided during the transport and handling of fresh MA containing fuels [2]. These challenges for fabrication, transport, and handling of MA fuels must be overcome in a dedicated MA fuel development program. The present work focuses mainly on the neutronic feasibility of MA transmutation as burnable poison in the VVER-1000 fuel assembly.

For the purpose of this study, the MA vector consisting of neptunium, americium, and curium from spent fuel of the VVER-440 [5] is adopted and given in Table 2. The parameters to be investigated are the k-inf of the fuel assembly versus burnup and the transmutation rates of MAs in the VVER-1000 LEU fuel assembly. The results are expected to reveal the MA transmutation possibility in the VVER-1000 LEU fuel assembly and the capability of MAs to substitute partially the gadolinium and boric acid in the VVER-1000 reactor.

3. Verification of the SRAC Model for the VVER-1000 LEU Fuel Assembly

The SRAC calculation model of the VVER-1000 LEU fuel assembly was verified against the MCNP6 calculations and the benchmark mean (BM) values [21]. The Monte Carlo calculations with the MCNP6 code using a modern nuclear data library were performed herein to support the verification of the SRAC deterministic model. In the SRAC calculations, the ENDF/B-VII.0 library that is the latest one

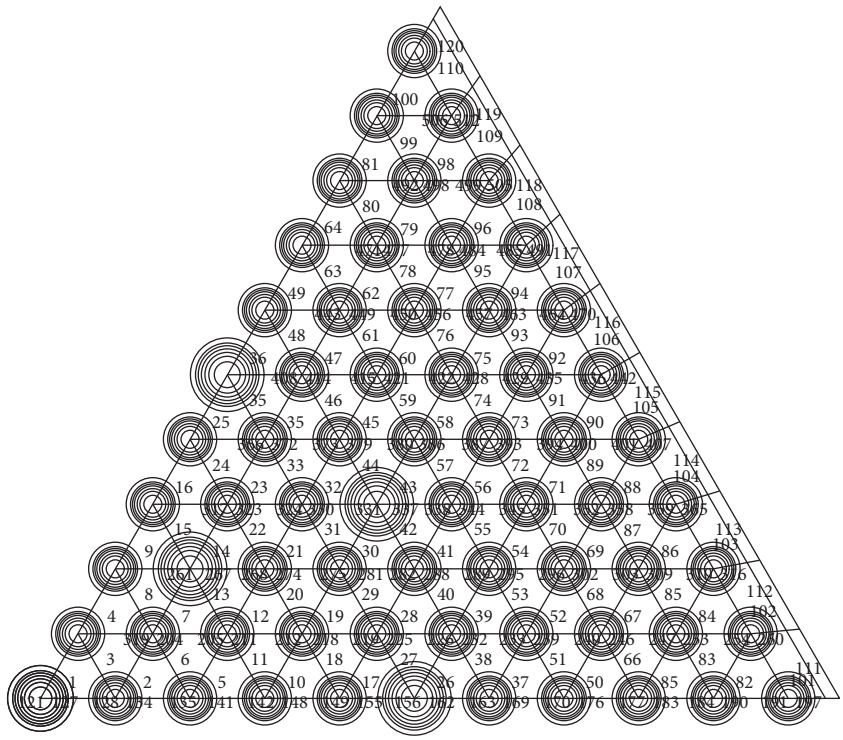

Figure 2: One-sixth model of the VVER-1000 LEU fuel assembly with SRAC.

Table 2: MA vector of VVER-440 spent fuel.

MA vector									
Isotope	^{237}Np	^{241}Am	^{242m}Am	^{243}Am	^{242}Cm	^{243}Cm	^{244}Cm	^{245}Cm	^{246}Cm
Fraction (at.%)	48.89	31.56	0.11	14.65	0.001	0.049	4.43	0.26	0.05

integrated with the SRAC code system was used whereas the ENDF/B-VII.1 library was utilized in the MCNP6 simulation. The burnup calculation for the VVER-1000 LEU fuel assembly was then performed with SRAC and MCNP6 under the operating poisoned condition (the S1 state) [21]. Under this condition, the fuel temperature is 1027 K and the moderator temperature is 575 K with equilibrium ^{135}Xe and ^{149}Sm concentrations and a power density of 108 MWt/m^3 up to a burnup of 40 MWd/kgHM. In both the SRAC and MCNP6 calculations, the UGD pins have been radially divided into five rings as required in the benchmark document [21] to account for the shielding effect due to the gadolinium isotopes and allow the calculation of nuclide concentrations as a function of the radial position in the UGD pins.

In the MCNP6 simulation, the statistical error of ~30 pcm was obtained as the neutron history of 5×10^6 for parallel depletion calculation was selected. Burnup calculations were performed with 160 steps of 0.25 MWd/kgHM. MCNP6 includes the new depletion capability linking steady state flux calculations in MCNP6 and nuclide depletion calculations in CINDER90 [23]. A steady state flux calculation is run to determine the system eigenvalue, group fluxes, energy integrated reaction rates, fission multiplicity, and recoverable energy per fission. CINDER90 then uses these values generated by MCNP6 to perform depletion calculation for number densities of the next burnup step. MCNP6 takes the new number densities generated by CINDER90 for the next steady state flux calculation. This linked process is repeated until the end of the final burnup step. However, the default nuclear data in MCNP6 are given at certain temperatures for heavy isotopes (293.6, 600, 900, 1200, and 2500 K) while the fuel temperature of 1027 K is needed in this calculation. There are various methods to cope with such kind of temperature dependence [25]. One of the suitable methods for MCNP6 is the on-the-fly (OTF) methodology for fitting of Doppler broadened cross sections and this method was applied in the present study. The OTF data for heavy isotopes in the VVER-1000 LEU fuel assembly that correspond to the temperature range of 293.6 to 1200 K were created from the ENDF/B-VII.1 library at the temperature of 293.6 K.

The PIJ module with its cell burnup routine of the SRAC code system [22] was utilized for the burnup calculation of the VVER-1000 LEU benchmark assembly. The PIJ module that is based on the collision probability method was used for lattice cell calculations. The cell burnup routine used one-group collapsed flux distribution and the collapsed microscopic cross sections to solve the depletion equation using Bateman's method. The burnup calculation using the cell burnup routine of the PIJ module was performed with 40 steps of 0.25 MWd/kgHM followed by 5 steps of 1.0 MWd/kgHM and 10 steps of 2.5 MWd/kgHM. The 107 neutron

energy groups based on the ENDF/B-VII.0 library were collapsed to four groups for use in the SRAC calculations.

The results for the k-inf as a function of burnup for the S1 state, i.e., the operating poisoned state, are shown in Figure 3. It can be seen that the k-inf calculated using SRAC agreed well with that calculated with MCNP6 and the BM value. Comparing with the BM values, the maximum differences of k-inf obtained from SRAC and MCNP6 calculations are 352 and 413 pcm, respectively, whereas those for other computational codes used in the benchmark such as MCU, TVS-M, WIMS8A, HELIOS, and MULTICELL are 440, 400, 460, 260, and 360 pcm, respectively [21]. The reactivity curve of the fuel assembly is relatively flat in the early burnup stage due to the use of Gd_2O_3 in the UGD pins for excess reactivity control. As the gadolinium isotopes burn out, the reactivity decreases with burnup in a nearly linear manner due to the effect of fissile material depletion and neutron absorber accumulation. It can also be seen that the effect of gadolinium depletion on the reactivity curve was well simulated by SRAC and MCNP6. The differences in the results obtained from SRAC and MCNP6 are attributed to the different transport and depletion calculation methods, the different nuclear data libraries used in SRAC and MCNP6, and the models using the two codes.

In addition, it was also confirmed that the nuclide concentrations for Cell 1 (UO_2 pin) and Cell 24 (UGD pin) as specified in the benchmark problem [21] as well as the radial nuclide concentrations for five rings of Cell 24 (UGD pin) calculated with SRAC and MCNP6 generally compare well with the BM values. Therefore, it is demonstrated that the SRAC calculation model for the VVER-1000 LEU fuel assembly developed in this study is reliable and it will be used for the investigation of MA transmutation possibility in the VVER-1000 LEU fuel assembly.

4. MA Transmutation in VVER-1000 LEU Fuel Assembly

4.1. Homogeneous Mixing of MAs in the UGD Pins. As the MAs are homogeneously mixed in the UGD pins of the VVER-1000 LEU fuel assembly, the gadolinium content and boron concentration were adjusted with varying content of MAs in order to maintain the reactivity of the fuel assembly. It is because the MAs can act as burnable poison and thus can partially replace functions of the gadolinium in the UGD pins and boric acid in the coolant [19, 20]. In this calculation, the content of MAs was loaded from 6 wt.% to 10 wt.%; the content of the gadolinium was reduced from 4 wt.% in the reference case to 2 wt.%, 2.5 wt.%, and 3 wt.% and the boron concentration was reduced correspondingly to compensate the negative reactivity insertion by the MAs. Table 3 summarizes the cases investigated here.

Although in the current practice the MA content in MA bearing fuels should be limited to a few percent (up to 5 wt.%) for the homogeneous case [2], the relatively high MA content from 6 wt.% to 10 wt.% was selected in this investigation to allow high MA loading amount so as to demonstrate a considerable MA transmutation rate from neutronic viewpoint. As the VVER-1000 LEU fuel assembly used in this study has 12 UGD pins, its configuration can be redesigned to accommodate more UGD pins (up to 36 pins) [26, 27]. With a larger number of the UGD pins, equivalent or even higher MA loading amount in the UGD pins as compared with this study can be easily obtained with the MA content not higher than 5 wt.%.

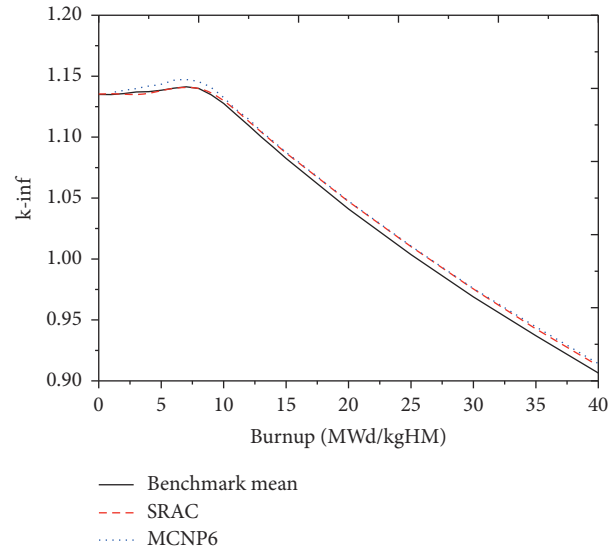

Figure 3: Infinite multiplication factor (k-inf) of the VVER-1000 LEU fuel assembly versus burnup obtained with SRAC and MCNP6 calculations.

Table 3: MA content, gadolinium content, and boron concentration used in this investigation.

MA content, wt.%	Gadolinium content, wt.%; Boron concentration, ppm		
6	2; 550	2.5; 500	3; 450
8	2; 500	2.5; 450	3; 400
10	2; 450	2.5; 400	3; 350

As shown in Table 3, the gadolinium content was first reduced to 2 wt.% and the boron concentration was decreased from 600 ppm (reference case) to 550 ppm, 500 ppm, and 450 ppm with respect to the MA content of 6 wt.%, 8 wt.%, and 10 wt.%. The results of the k-inf of the VVER-1000 LEU fuel assembly versus burnup were displayed in Figures 4–6 for the cases when reducing the gadolinium content to 2 wt.% and loading the MA content of 6, 8, and 10 wt.%. It was found that the fuel cycle length when loading MAs from 6 to 10 wt.% and decreasing the gadolinium content to 2 wt.% was substantially reduced as compared to the reference case. As can be seen in Figures 4–6, the combined reduction of gadolinium content to 2 wt.% and respective boron concentration could lead to a cycle length comparable to the reference case. However, the excess reactivity in these cases was generally higher at the early burnup steps and became smaller than the reference case after about 7 MWd/kgHM as gadolinium burned out.

The gadolinium content was therefore increased from 2 to 2.5 wt.% to expect a decrease of the aforementioned high

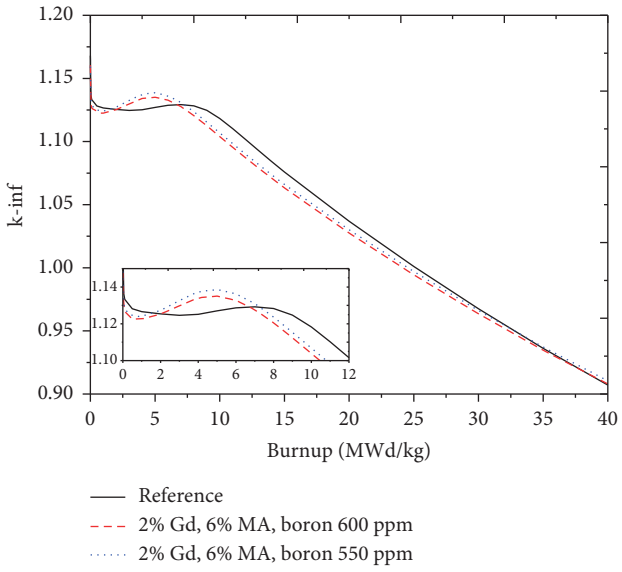

Figure 4: The k-inf versus burnup when loading 6 wt.% of MAs and reducing GD to 2 wt.%.

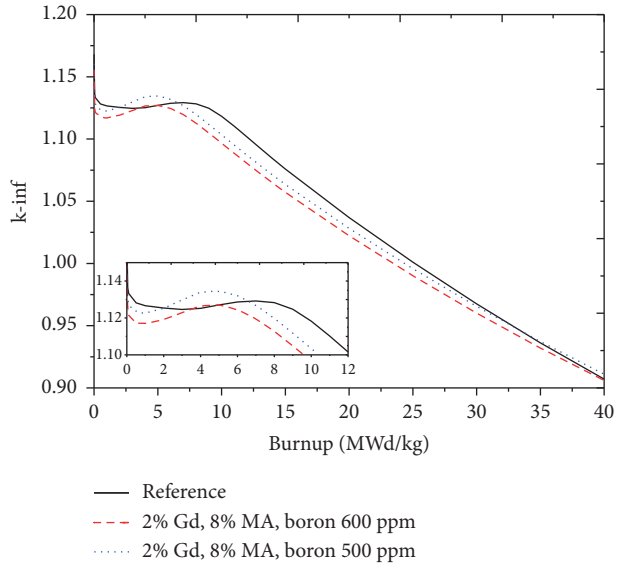

Figure 5: The k-inf versus burnup when loading 8 wt.% of MAs and reducing GD to 2 wt.%.

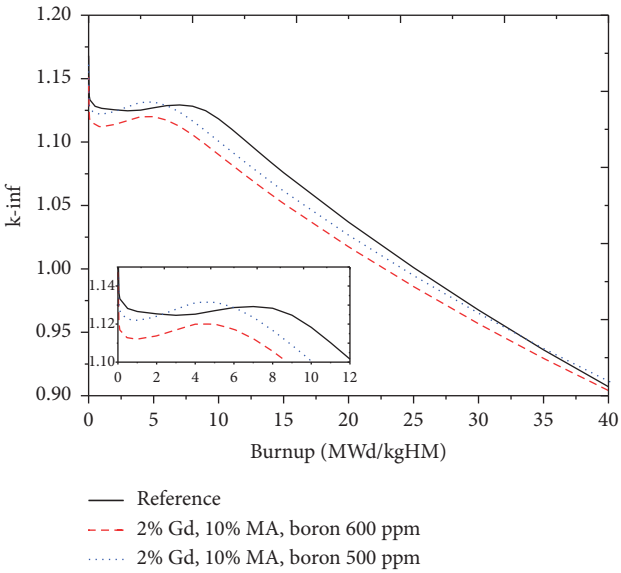

Figure 6: The k-inf versus burnup when loading 10 wt.% of MAs and reducing GD to 2 wt.%.

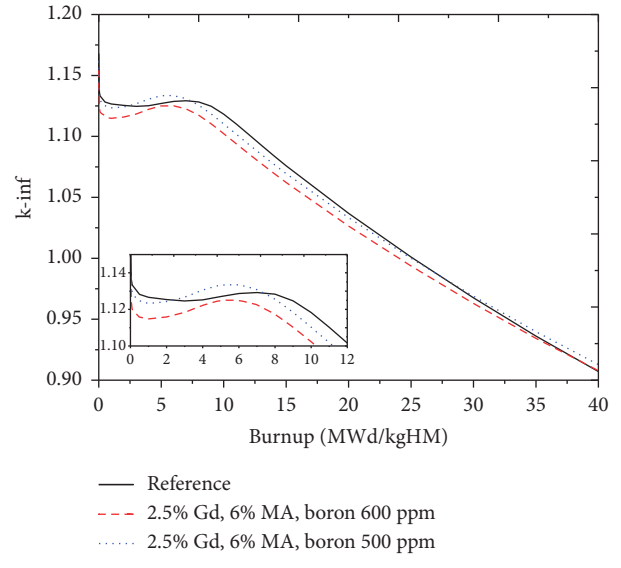

Figure 7: The k-inf versus burnup when loading 6 wt.% of MAs and reducing GD to 2.5 wt.%.

excess reactivity at the early burnup steps and the boron concentration was adjusted to 500 ppm, 450 ppm, and 400 ppm with respect to the MA content of 6 wt.%, 8 wt.%, and 10 wt.%. Figures 7–9 show that the behavior of the k-inf versus burnup in the case of reducing the gadolinium content to 2.5 wt.% was somewhat different from that with the gadolinium content of 2 wt.%. Namely, the excess reactivity in the cases of reducing only the gadolinium content to 2.5 wt.% was smaller, leading to smaller cycle lengths. To overcome this disadvantage, reducing the boron concentration to 500 ppm, 450 ppm, and 400 ppm with respect to the MA content of 6 wt.%, 8 wt.%, and 10 wt.% could lead to a comparable cycle length while still keeping the excess reactivity equivalent or even somewhat lower than the reference case as can be seen in Figures 7–9.

The gadolinium content was further increased from 2.5 to 3 wt.% and the boron concentration was adjusted to 450 ppm, 400 ppm, and 350 ppm with respect to the MAs content of 6 wt.%, 8 wt.%, and 10 wt.%. It was found that the behavior of the k-inf versus burnup in these cases (see Figures 10–12) is very similar to that with the gadolinium content of 2.5 wt.% as mentioned previously. Nonetheless, the cycle length when loading 6 wt.%, 8 wt.%, and 10 wt.% of MAs with the gadolinium content of 3 wt.% was further improved and became almost identical to the reference case as the boron concentration was reduced to 450 ppm,

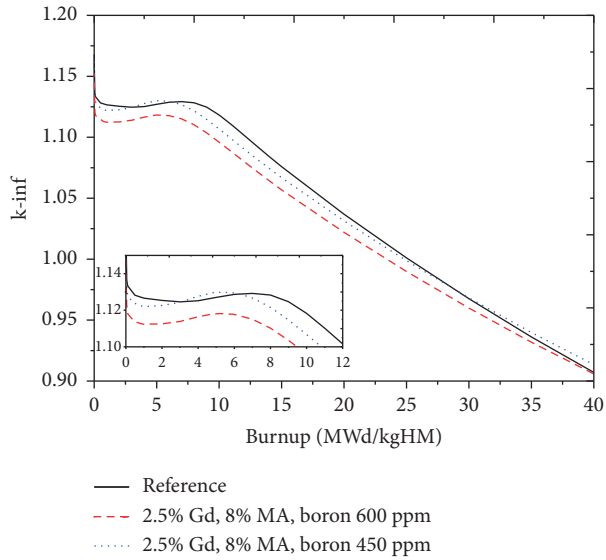

Figure 8: The k-inf versus burnup when loading 8 wt.% of MAs and reducing GD to 2.5 wt.%.

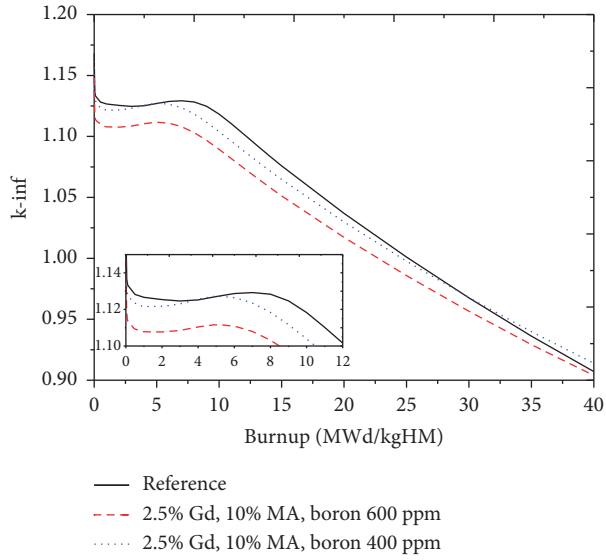

Figure 9: The k-inf versus burnup when loading 10 wt.% of MAs and reducing GD to 2.5 wt.%.

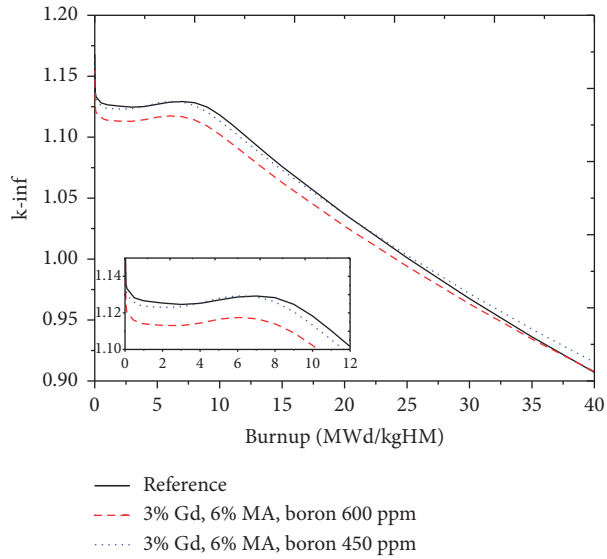

Figure 10: The k-inf versus burnup when loading 6 wt.% of MAs and reducing GD to 3 wt.%.

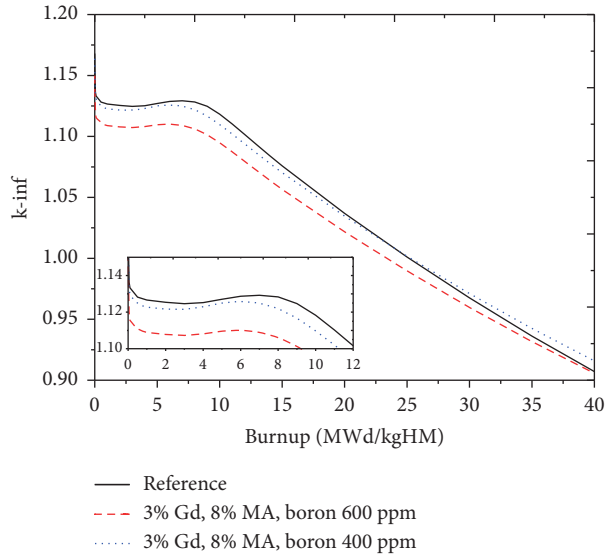

Figure 11: The k-inf versus burnup when loading 8 wt.% of MAs and reducing GD to 3 wt.%.

400 ppm, and 350 ppm, respectively, as shown in Figures 10–12. Moreover, the excess reactivity at the beginning of the cycle was also generally reduced in comparison to the reference case.

The results illustrated in Figures 4–12 also imply that the MAs with the content of up to 10 wt.% can be loaded into the VVER-1000 LEU fuel assembly without significantly affecting the fuel cycle length by means of reducing the gadolinium content and the boron concentration to offset the negative reactivity insertion by the MAs. For the MA loading from 8 to 10 wt.%, it was found that the lower excess reactivity and equivalent cycle length as compared to the reference case can be obtained with the gadolinium content being reduced to around 2.5–3.0 wt.% and the boron concentration being reduced to around 350–400 ppm. As a result, loading 10 wt.% of MAs into the UGD pins is recommended for the sake of excess reactivity control and high loading amount of MAs while keeping almost the same cycle length with the reference case.

The transmutation of the MA isotopes is shown in Figures 13–17 for the cases when loading 10 wt.% of MAs and adjusting the gadolinium content and boron concentration. As can be seen in these figures, the concentrations of ^{237}Np, ^{241}Am, and ^{243}Am decreased with fuel burnup while those of ^{244}Cm and ^{245}Cm accumulated with fuel burnup. Figures 13–17 also indicate that the transmutation of the MA

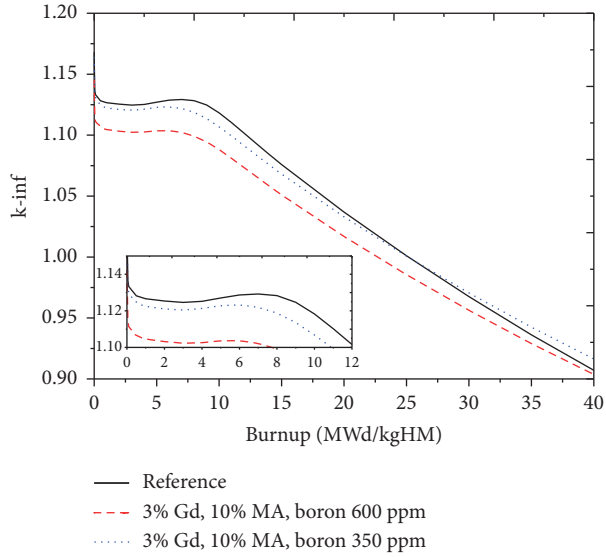

Figure 12: The k-inf versus burnup when loading 10 wt.% of MAs and reducing GD to 3 wt.%.

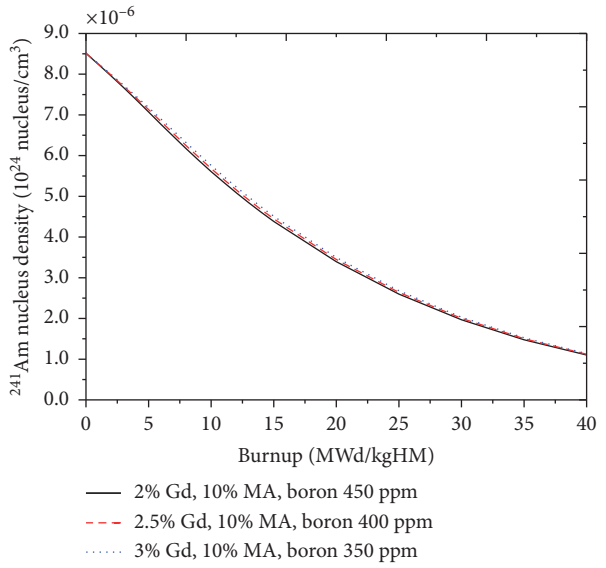

Figure 14: Transmutation of ^{241}Am versus burnup when loading 10 wt.% of MAs.

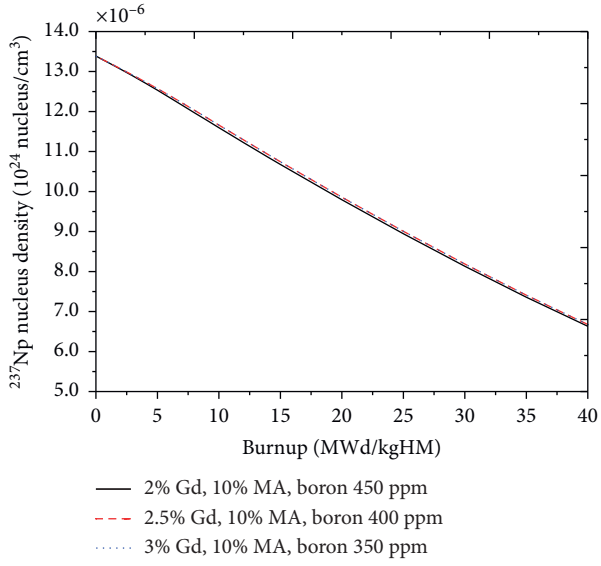

Figure 13: Transmutation of ^{237}Np versus burnup when loading 10 wt.% of MAs.

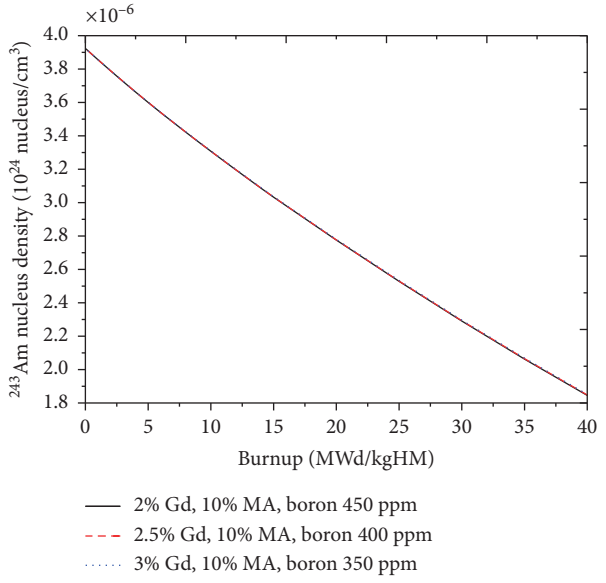

Figure 15: Transmutation of ^{243}Am versus burnup when loading 10 wt.% of MAs.

isotopes versus burnup slightly depends on the adjustments of the gadolinium content and boron concentration. After 306 days, the ^{237}Np concentration reduced to ~15.63%, the ^{241}Am concentration reduced to ~38.58%, and the ^{243}Am concentration reduced to ~18.48%, whereas those of ^{244}Cm and ^{245}Cm increased to ~51.60% and ~103.13% as illustrated in Table 4 when loading 10 wt.% of MAs and reducing the gadolinium content to 3 wt.% and boron concentration to 350 ppm. Hence, ^{237}Np, ^{241}Am, and ^{243}Am can be significantly transmuted with a transmutation rate as high as 38.58% for ^{241}Am. Nevertheless it is noticed that ^{244}Cm and ^{245}Cm accumulate with high rates though their concentrations are relatively small. This issue was also reported in recent publications for thermal reactors [2, 20]. It is recalled that the radiotoxicity of MAs in spent fuel from power reactors is mainly contributed by ^{241}Am (half-life of 432.2 years) and ^{244}Cm (half-life of 18.1 years). If actinides from PWR-type reactors over 100 years of storage are considered, the radiotoxicity of ^{241}Am contributes more than 90% of the total radiotoxicity while the contribution from other actinides is less than 10% [18]. Hence, transmutation of ^{241}Am could contribute to a significant reduction of the radiotoxicity level of the long-lived radioactive waste.

The results demonstrate that the transmutation of MAs recycled from spent nuclear fuel in the VVER-1000 fuel assembly is feasible from neutronic viewpoint and the

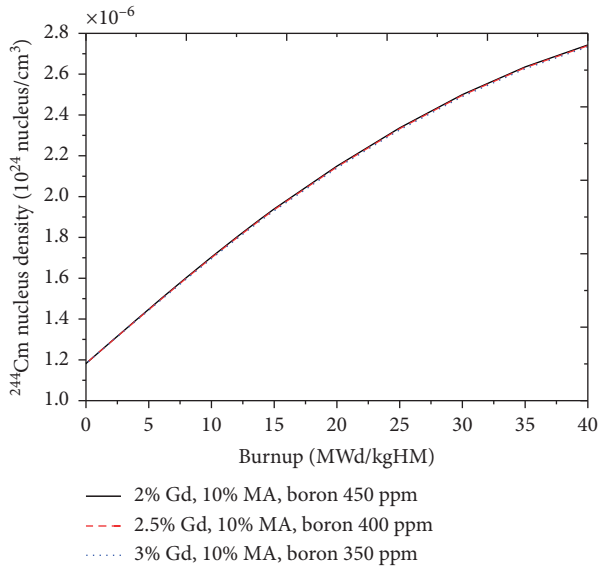

Figure 16: Transmutation of ^{244}Cm versus burnup when loading 10 wt.% of MAs.

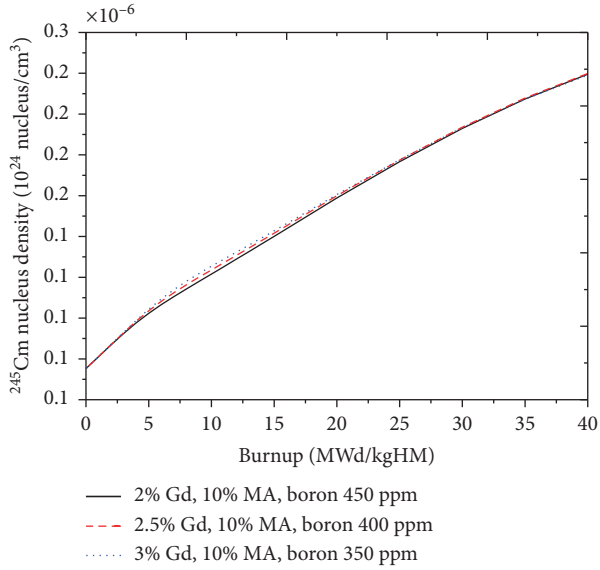

Figure 17: Transmutation of ^{245}Cm versus burnup when loading 10 wt.% of MAs.

total transmutation rate of ~20% can be achieved. Also, the role of MAs as burnable absorbers in partial replacement to the gadolinium and boric acid in the VVER-1000 reactor was confirmed and in line with recent publications for PWRs [19, 20]. It is worth noting that the reduction of the boron concentration not only could help attain a more negative moderator temperature coefficient during the core lifetime but also could allow minimizing the operation of a complicated chemical control system as well as reducing the corrosion of structural materials [26]. Furthermore, the reduced gadolinium content could help improve the thermal conductivity and melting point of the UGD pins [28].

Table 4: Transmutation rate in case of homogeneous loading with 10 wt.% of MAs.

Isotope	Initial amount (g)	Residual amount at 306 days (g)	Mass reduced after 306 days (g)	Disappearance rate after 306 days (%)
^{237}Np	896.78	756.59	140.19	15.63
^{241}Am	580.05	356.29	223.76	38.58
^{243}Am	269.52	219.72	49.80	18.48
^{244}Cm	81.54	123.62	−42.08	−51.60
^{245}Cm	4.79	9.73	−4.94	−103.13
Total	1832.67	1465.94	366.73	20.01

4.2. Coating a Thin Layer of MAs to the UGD Pins. In addition to homogeneous mixing of MAs in the UGD pins as above, the heterogeneous loading of MAs in the UGD pins of the VVER-1000 LEU fuel assembly was also considered herein. The MAs were coated as a thin layer at the outside of the UGD pellets as shown in Figure 18. The thickness of the cladding was kept untouched and the outer radius of the UGD region was reduced to accommodate the layer of MAs. For the purpose of MA burning, the MA content of 10 wt.% was selected in this investigation. The MA-coated layer (see Figure 18) equivalent to homogeneous loading with 10 wt.% of MAs is 0.01981 cm thick. Similar to the case of homogeneous mixing, the gadolinium content and boron concentration were also reduced to compensate the negative reactivity insertion by the MAs.

The results of the k-inf of the VVER-1000 LEU assembly versus burnup when coating MAs to the UGD pins and reducing the gadolinium content and boron concentration are shown in Figures 19–21 in relation to the reference case. It can be seen that the cases of reducing only the gadolinium content led to a significantly lower excess reactivity at the beginning of the cycle and a considerably shorter cycle length. This behavior of the k-inf versus burnup is very similar to that shown in Figures 6, 9, and 12 for the cases of homogeneous loading. Hence, the boron concentration was again reduced to 450 ppm, 400 ppm, and 350 ppm with respect to the gadolinium content of 2 wt.%, 2.5 wt.%, and 3 wt.%. Figures 19–21 show that the excess reactivity at the early burnup steps when reducing both the gadolinium content and boron concentration was smaller than or comparable to the reference case. Nevertheless, the cycle length when reducing the boron concentration to 350 ppm became identical to the reference case, while that with the boron concentration of 400 ppm and 450 ppm was somewhat shorter. Consequently, reducing the gadolinium content to 3 wt.% and boron concentration to 350 ppm is recommended when coating with 10 wt.% of MAs to the UGD pellets.

The transmutation of the MA isotopes when coating with 10 wt.% of MAs and reducing the gadolinium content to 3 wt.% and boron concentration to 350 ppm is given in Table 5. Comparing the results shown in Tables 4 and 5 shows that the difference in the transmutation rate of the MA isotopes between homogeneous and heterogeneous loadings was relatively small. However, the transmutation mass in the case of heterogeneous loading was ~6.8% higher

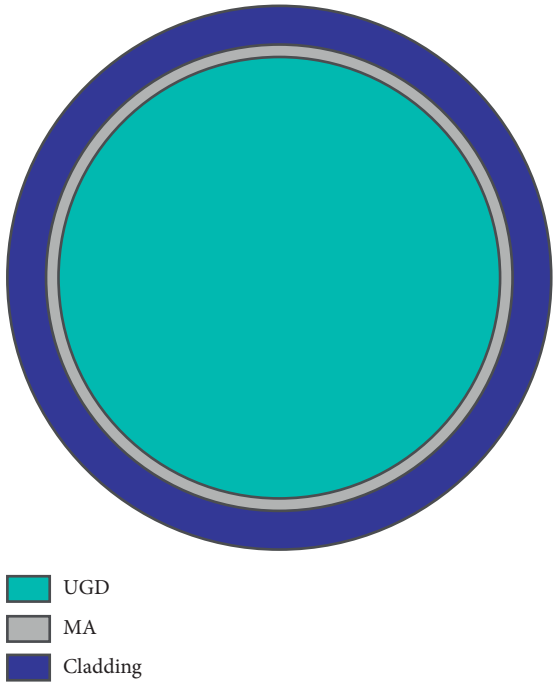

Figure 18: Coating a thin layer of MAs to the UGD pellet.

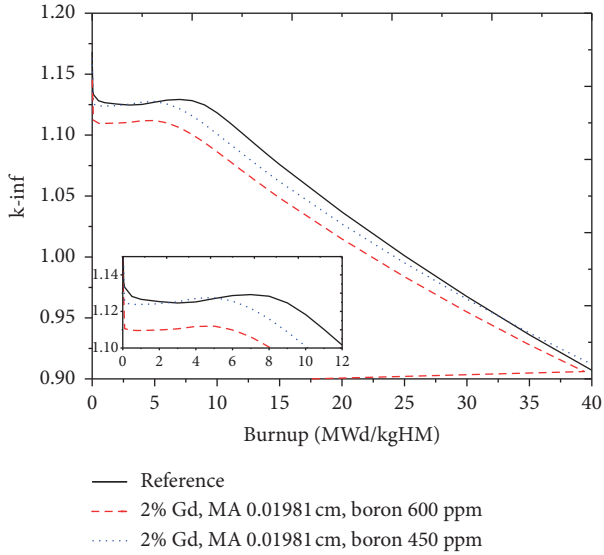

Figure 19: The k-inf versus burnup when coating a thin layer of MAs to the UGD pins and reducing GD to 2 wt.%.

than that of homogeneous loading. It is because the spatial self-shielding effect due to the heterogeneous loading of MAs will affect the burnup of the fuel and the transmutation of MAs [20]. For the case of heterogeneous loading in this study, the core neutrons reach the MA layer firstly and the MA layer is thin enough, leading to such increase in the transmutation of MAs as compared to the case of homogeneous mixing. Furthermore, the spatial self-shielding effect also affected the depletion of ^{155}Gd and ^{157}Gd as illustrated in Figures 22 and 23, which show that the

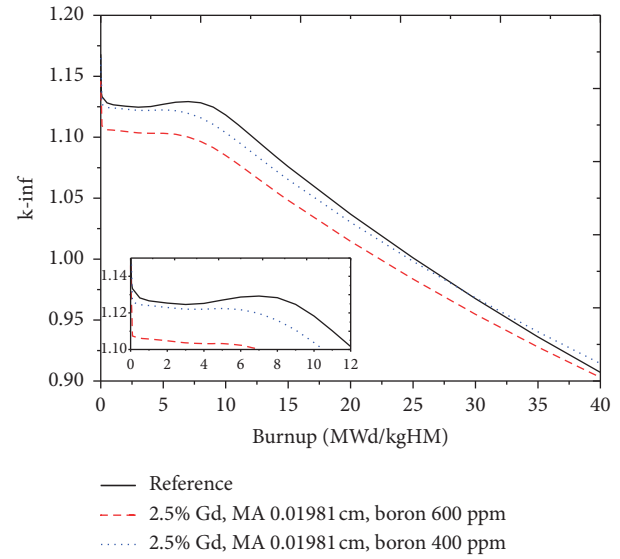

Figure 20: The k-inf versus burnup when coating a thin layer of MAs to the UGD pins and reducing GD to 2.5 wt.%.

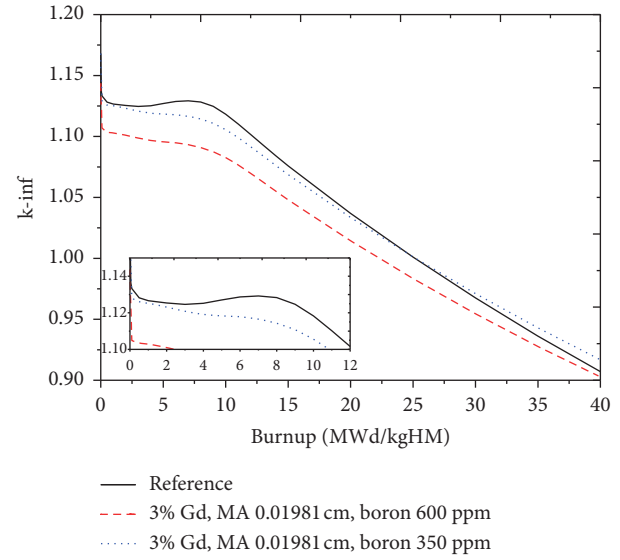

Figure 21: The k-inf versus burnup when coating a thin layer of MAs to the UGD pins and reducing GD to 3.0 wt.%.

Table 5: Transmutation rate in case of heterogeneous loading with 10 wt.% of MAs.

Isotope	Initial amount (g)	Residual amount after 306 days (g)	Mass reduced after 306 days (g)	Disappearance rate after 306 days (%)
^{237}Np	896.78	746.44	150.34	16.76
^{241}Am	580.05	341.73	238.32	41.09
^{243}Am	269.52	218.40	51.12	18.97
^{244}Cm	81.54	122.82	−41.28	−50.63
^{245}Cm	4.79	11.60	−6.81	−142.17
Total	1832.67	1440.98	391.69	21.37

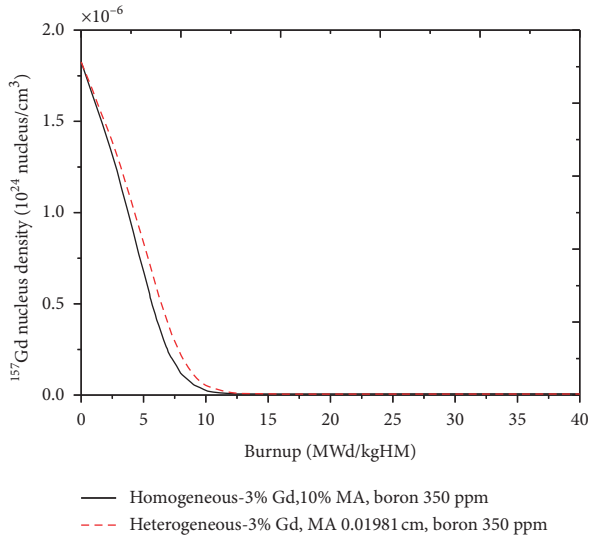

Figure 22: Comparison of the depletion of ^{155}Gd for two MA loading models.

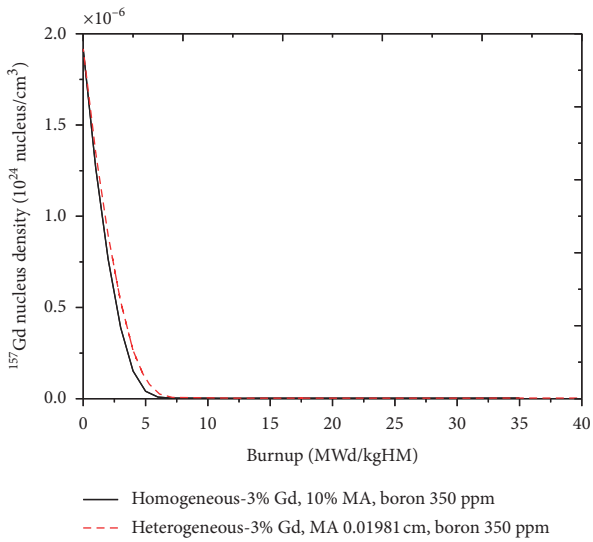

Figure 23: Comparison of the depletion of ^{157}Gd for two MA loading models.

gadolinium isotopes depleted somewhat slower in the case of heterogeneous loading.

5. Conclusions

The possibility of MA transmutation as burnable poison in the VVER-1000 LEU fuel assembly was examined using the SRAC code system. The SRAC calculation model for the VVER-1000 LEU fuel assembly was verified against the MCNP6 calculations and the available published benchmark results. Two models of MA loading were considered: homogeneous mixture in the UGD pellet and heterogeneous coated layer around the UGD pellet. The gadolinium content and the boron concentration were reduced correspondingly to compensate the negative reactivity insertion by MA loading.

It was found that the MAs can be loaded up to 10 wt.% into the UGD pins and the combined reduction in the gadolinium content and boron concentration could help facilitate the excess reactivity control at the beginning of the fuel cycle without significant effect on the cycle length. As in the current practice the MA content in MA bearing fuels should be limited to a few percent (up to 5 wt.%) for the homogeneous case [2], the limitations regarding the relatively high MA content from 6 wt.% to 10 wt.% can be possibly overcome by increasing the number of the UGD pins (up to 36 pins) in the VVER-1000 fuel assembly [26, 27]. Increasing the number of the UGD pins could reduce the MA content in the UGD pins less than 5 wt.% while allowing a high total MA loading amount in the whole assembly and thus avoid serious shielding issues in the fabrication, transport, and handling of MA bearing fuels.

For both cases of homogeneous and heterogeneous loadings, the total transmutation rate of ~20% could be obtained. However, the comparison between the two cases shows that the transmutation mass could be increased by ~6.8% for the case of coating a thin layer of MA to the UGD pins. The results show that ^{237}Np, ^{241}Am, and ^{243}Am can be significantly transmuted with a transmutation rate as high as ~40% for ^{241}Am. This advantage is somewhat offset by the unfavorable accumulation of ^{244}Cm and ^{245}Cm at high rates with fuel burnup. However, if actinides from PWR-type reactors over 100 years of storage are considered, more than 90% of the total radiotoxicity is contributed by ^{241}Am [18]. Therefore, transmuting ^{241}Am could lead to a significant decrease of the total long-term radiotoxicity of MAs.

Consequently, it is highly recommended that transmutation of MAs as burnable poison in the VVER-1000 reactor is feasible taking into account the fact that the excess reactivity control and the inherent safety characteristics of the VVER-1000 reactor can be further improved as MAs can partially replace the gadolinium and boric acid. Further investigation on transmutation of MAs at a full core level and MOX core of the VVER-1000 reactor when coating a thin layer of MA to the UGD pins is being planned.

Data Availability

Data will be available upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

This research was funded by the Ministry of Science and Technology of Vietnam under Grant no. DTCB.04/17/VKHKTHN.

References

[1] International Atomic Energy Agency, "Status of minor actinide fuel development," IAEA Nuclear Energy Series, Vienna, Austria, 2009.

[2] OECD/NEA, *Minor Actinide Burning in Thermal Reactors*, Nuclear Energy Agency, Paris, France, 2013.

[3] R. Jubin, "Spent fuel reprocessing, introduction to nuclear chemistry and fuel cycle separations course, consortium for risk evaluation with stakeholder participation," 2008, http://www.cresp.org/education/courses/shortcourse/.

[4] C. H. M. Broeders, E. Kiefhaber, and H. W. Wiese, "Burning transuranium isotopes in thermal and fast reactors," *Nuclear Engineering and Design*, vol. 202, no. 2-3, pp. 157–172, 2000.

[5] Z. Perkó, J. L. Kloosterman, and S. Fehér, "Minor actinide transmutation in GFR600," *Nuclear Technology*, vol. 177, no. 1, pp. 83–97, 2012.

[6] T. Kooyman, L. Buiron, and G. Rimpault, "A comparison of curium, neptunium and americium transmutation feasibility," *Annals of Nuclear Energy*, vol. 112, pp. 748–758, 2018.

[7] H. N. Tran and Y. Kato, "New^{237}Np burning strategy in a supercritical CO_2-cooled fast reactor core attaining zero burnup reactivity loss," *Nuclear Science and Engineering*, vol. 159, no. 1, pp. 83–93, 2008.

[8] H. N. Tran, Y. Kato, P. H. Liem, V. K. Hoang, and S. M. T. Hoang, "Minor actinide transmutation in supercritical-CO_2-cooled and sodium-cooled fast reactors with low burnup reactivity swings," *Nuclear Technology*, vol. 205, no. 11, pp. 1460–1473, 2019.

[9] N. Takaki, "Neutronic potential of water cooled reactor with actinide closed fuel cycle," *Progress in Nuclear Energy*, vol. 37, no. 1–4, pp. 223–228, 2000.

[10] E. Shwageraus, P. Hejzlar, and M. S. Kazimi, "A combined nonfertile and UO_2 PWR fuel assembly for actinide waste minimization," *Nuclear Technology*, vol. 149, no. 3, pp. 281–303, 2005.

[11] T. A. Taiwo, T. K. Kim, J. A. Stillman, R. N. Hill, M. Salvatores, and P. J. Finck, "Assessment of a heterogeneous PWR assembly for plutonium and minor actinide recycle," *Nuclear Technology*, vol. 155, no. 1, pp. 34–54, 2006.

[12] D. Tincher and T. W. Knight, "Feasibility study of minor actinide transmutation in light water reactors with various Am/Cm separation efficiencies," *Nuclear Engineering and Design*, vol. 241, no. 12, pp. 5295–5307, 2011.

[13] M. A. Pope, R. S. Sen, A. M. Ougouag, G. Youinou, and B. Boer, "Neutronic analysis of the burning of transuranics in fully ceramic micro-encapsulated tri-isotropic particle-fuel in a PWR," *Nuclear Engineering and Design*, vol. 252, pp. 215–225, 2012.

[14] B. Liu, K. Wang, J. Tu, F. Liu, L. Huang, and W. Hu, "Transmutation of minor actinides in the pressurized water reactors," *Annals of Nuclear Energy*, vol. 64, pp. 86–92, 2014.

[15] A.-A. Zakari-Issoufou, X. Doligez, A. Somaini et al., "Americium mono-recycling in PWR: a step towards transmutation," *Annals of Nuclear Energy*, vol. 102, pp. 220–230, 2017.

[16] B. Liu, R. Jia, R. Han, X. Lyu, J. Han, and W. Li, "Minor actinide transmutation characteristics in AP1000," *Annals of Nuclear Energy*, vol. 115, pp. 116–125, 2018.

[17] V. Sebian, V. Necas, and P. Darilek, "Transmutation of spent fuel in reactor VVER-440," *Journal of Electrical Engineering*, vol. 52, no. 9-10, pp. 299–302, 2001.

[18] B. R. Bergelson, A. S. Gerasimov, and G. V. Tikhomirov, "Transmutation of actinides in power reactors," *Radiation Protection Dosimetry*, vol. 116, no. 1–4, pp. 675–678, 2005.

[19] W. Hu, B. Liu, X. Ouyang et al., "Minor actinide transmutation on PWR burnable poison rods," *Annals of Nuclear Energy*, vol. 77, pp. 74–82, 2015.

[20] W. Hu, J. Jing, J. Bi, C. Zhao, B. Liu, and X. Ouyang, "Minor actinides transmutation on pressurized water reactor burnable poison rods," *Annals of Nuclear Energy*, vol. 110, pp. 222–229, 2017.

[21] OECD/NEA, *A VVER-1000 LEU and MOX Assembly ComputationalBenchmark*, vol. 10, Nuclear Energy Agency, NEA/NSC/DOC, Paris, France, 2002.

[22] K. Okumura, T. Kugo, K. Kaneko, and K. Tsuchihashi, *SRAC2006: A Comprehensive Neutronics Calculation Code System*, JAEA-Data/Code 2007-004, Tokai, Japan, 2007.

[23] D. B. Pelowitz, *MCNP6TM User's Manual*, Los Alamos National Laboratory, Santa Fe, NM, USA, 2013.

[24] A. Kotchetkov, I. Krivitskiy, N. Rabotnov, A. Tsiboulia, and S. Iougai, "Calculation and experimental studies on minor actinides reactor transmutation," in *Proceedings of the Fifth OECD/NEA Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation*, pp. 289–303, Mol, Belgium, November 1998.

[25] F. Brown, W. Martin, G. Yesilyurt, and S. Wilderman, *On-The-Fly Neutron Doppler Broadening for MCNP*, Los Alamos National Laboratory, Santa Fe, NM, USA, 2012.

[26] H. N. Tran, V. K. Hoang, P. H. Liem, and H. T. P. Hoang, "Neutronics design of VVER-1000 fuel assembly with burnable poison particles," *Nuclear Engineering and Technology*, vol. 51, no. 7, pp. 1729–1737, 2019.

[27] L. Frybortova, "Recommended strategy and limitations of burnable absorbers used in VVER fuel assemblies," *Nuclear Science and Techniques*, vol. 30, no. 8, p. 129, 2019.

[28] P. Cudrnak and V. Necas, "Optimization of fuel assembly with gadolinium for LWRS," *Journal of Power Technologies*, vol. 91, no. 4, pp. 186–190, 2011.