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Study on transmutation efficiency of the VVER-1000 fuel assembly with different minor actinide compositions

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Abstract: The feasibility of transmutation of minor actinides recycled from the spent nuclear fuel in the VVER-1000 LEU (low enriched uranium) fuel assembly as burnable poison was examined in our previous study. However, only the minor actinide vector of the VVER-440 spent fuel was considered. In this paper, various vectors of minor actinides recycled from the spent fuel of VVER-440, PWR-1000, and VVER-1000 reactors were therefore employed in the analysis in order to investigate the minor actinide transmutation efficiency of the VVER-1000 fuel assembly with different minor actinide compositions. The comparative analysis was conducted for the two models of minor actinide loading in the LEU fuel assembly: homogeneous mixing in the UGD (Uranium-Gadolinium) pins and coating a thin layer to the UGD pins. The parameters to be analysed and compared include the reactivity of the LEU fuel assembly versus burnup and the transmutation of minor actinide nuclides when loading different minor actinide vectors into the LEU fuel assembly.

Keywords: *VVER-1000 LEU fuel assembly, burnable poison, minor actinide transmutation*.

I. INTRODUCTION

The radioactive waste and spent nuclear fuel discharged from nuclear power plants causes a big issue for the countries holding such nuclear installations. It is widely recognized that a light water reactor (LWR) with electric capacity of 1000 MWe, on average, produces 20-30 metric tonnes of spent nuclear fuel annually, which consist of approximately 95 wt% uranium, 1 wt% plutonium, 4 wt% fission products and minor actinide (MA) [1]. In the spent fuel, only with a very small amount of MAs, they still 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 as well as to reduce its cumulative radiotoxicity to the environment, separation and transmutation of the plutonium and MAs in the used fuel are esssential [2]. It has been realized that the transmutation of these actinide into either short-lived fission products or valued fissile or stable isotopes can be accomplished in fast reactors, subcritical reactors or thermal reactors [1,3-7].

The VVER-1000 reactor (the Russian Pressurized Water Reactor, PWR) is nowadays operated in various East European and Asian countries [8,9]. In addition to the Western PWRs that have been extensively studied for their MA

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transmutation capabilities [10-14], the VVER-1000 is also considered as a potential candidate for transmutation of actinide 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 the MAs in the burnable poison rods [15,16] or in some other locations in the PWR fuel assemblies has been found technically feasible and recommended as potential transmutation methods for LWRs, in particular the unique advantage of loading MAs to partially replace the excess reactivity control functions of gadolinium and boric acid.

In a previous study [17], the feasibility of MA transmutation in VVER-1000 LEU fuel assembly [18] as burnable poison was studied and the results showed that the total MA transmutation rate of ~20% could be obtained. However, only the MA vector of the VVER-440 spent fuel was considered. In the present work, different MA compositions recycled from the spent fuels of VVER-440 [4], PWR-1000 [15] and VVER-1000 [19] with different burnup levels and cooling time were therefore employed in the analysis in order to estimate the effects of various MA contents in the spent fuel to the infinite multiplication factor (k-inf) of the VVER-1000 LEU fuel assembly versus burnup as well as the MA transmutation efficiency. The SRAC code [20] was used for modeling of the VVER-1000 LEU fuel assembly based on the ENDF/B-VII.0 library. The comparative analysis was conducted for the two models of MA loading in the LEU fuel assembly: homogeneous mixing in the UGD (Uranium-Gadolinium) pins and coating a thin layer to the UGD pins. The MA loading into the LEU fuel assembly will be performed without significant modification of the assembly configuration to minimize the cost for fuel fabrication process and respective changes in reactor core design. The constraint for these MA loadings is to ensure insignificant change in the reactivity of the fuel assembly while providing considerable MA transmutation rates.

II. CALCULATION METHOD

The VVER-1000 LEU fuel assembly specified in the OECD VVER-1000 LEU and MOX (mixed oxide) Assembly Computational Benchmark [18] is utilized in the present investigation to examine the possibility of MA transmutation as burnable poison in the VVER-1000 reactor. The configuration of the LEU fuel assembly are shown in Fig. 1. The LEU assembly consists of 300 fuel pin cells with 3.7wt% ^{235}U, 12 UGD pin cells with 3.6wt% ^{235}U and 4wt% Gd_2O_3, 18 water filled guide tubes for control insertion and one central water filled instrumentation tube. The LEU fuel assembly is modeled by the SRAC code. The one-sixth of the LEU fuel assembly modeled by the PIJ module of SRAC is presented in Fig. 2; the burnup calculations were performed with the BURN-UP module of SRAC; and the 107 energy groups based on the ENDF/B-VII.0 nuclear data library were used in the SRAC calculations.

In this investigation, we intend to load the MAs in the UGD pins of the 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 pins; and (2) coating a thin layer of MAs around the UGD pellets. In these cases, different vectors of MAs were

employed including those recycled from the spent fuel of VVER-440 with 45 GWd/tonne burnup and 5 years of cooling [4], PWR-1000 with 33 MWd/tonne burnup and 10 years cooling [15] and VVER-1000 reactors with 40 GWd/tonne burnup and 10 years of cooling [19]. The MA vectors recycled from the spent fuels of the VVER-440, PWR-1000 and VVER-1000 are given in Table I. The parameters to be investigated are the k-inf of the LEU fuel assembly versus burnup and the MA transmutation rates in the LEU fuel assembly.

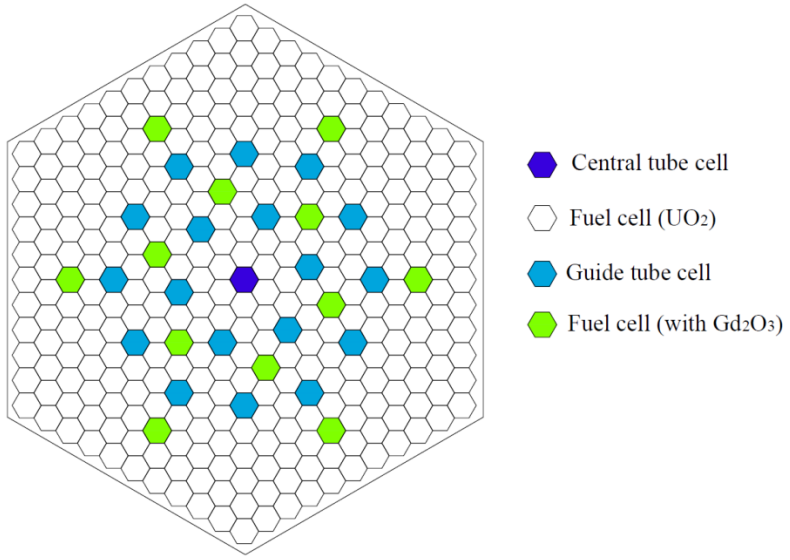

Fig. 1. Configuration of the VVER-1000 LEU fuel assembly

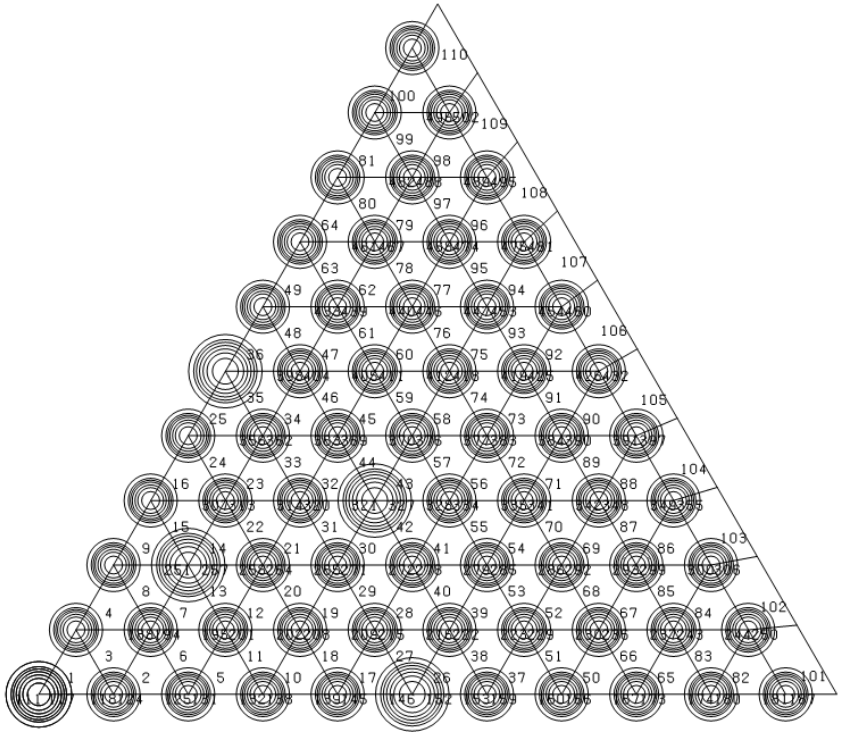

Fig. 2. One-sixth model of the VVER-1000 LEU fuel assembly by SRAC

Table I. MA vectors used in the analysis

MA vector (Fraction - at.%)									
Isotope	^{237}Np	^{241}Am	^{242m}Am	^{243}Am	^{242}Cm	^{243}Cm	^{244}Cm	^{245}Cm	^{246}Cm
VVER-440	48.89	31.56	0.11	14.65	0.001	0.049	4.43	0.26	0.05
PWR-1000	41.80	47.86	0.0	8.62	0.0	0.0	1.63	0.09	0.0
VVER-1000	0.0	83.75	0.10	13.16	1.22×10^{-6}	0.03	2.73	0.23	3.59×10^{-6}

III. MA TRANSMUTATION IN THE VVER-1000 FUEL ASSEMBLY

A. 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 the functions of the gadolinium in the UGD pins and boric acid in the coolant for excess reactivity control of the fuel assembly [15,16]. In this calculation, the content of MAs was loaded up to 10 wt%; the content of the gadolinium was reduced from 4.0 wt% in the reference case to 2 wt%, 2.5 wt%, 3 wt% and the boron concentration was reduced correspondingly to compensate the negative reactivity insertion by the MAs.

The results of the k-inf of the VVER-1000 LEU fuel assembly versus burnup were illustrated in Fig. 3 for cases with MA content of 10 wt%. The gadolinium content was first reduced to 2 wt% and the boron concentration was decreased from 600 ppm (reference case) to 450 ppm with respect to the MA content of 10 wt%. It was found that the fuel cycle length when loading 10 wt% of MAs and decreasing the gadolinium content to 2 wt% and the boron concentration to 450 ppm was substantially reduced as compared to the reference case. Additionally, in the case of loading MAs recycled from spent fuel of VVER-440 reactor, the excess reactivity 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 excess reactivity at the early burnup steps and the boron concentration was adjusted to 400 ppm with respect to the MA content of 10 wt%. As can be seen in Fig. 3, adjusting the gadolinium content to 2.5 wt% and the boron concentration to 400 ppm could lead to a comparable cycle length while still keeping the excess reactivity somewhat lower than the reference case.

The gadolinium content was further increased from 2.5 to 3 wt% and the boron concentration was adjusted to 350 ppm with respect to the MAs content of 10 wt%. It was found that the behaviour of the k-inf versus burnup in these cases is very similar to those with the gadolinium content of 2.5 wt% as previously mentioned. However, the cycle length when loading 10 wt% of MA with the gadolinium content of 3 wt% and boron concentration of 350 ppm was further improved and became almost identical to the reference case. Moreover, the excess reactivity of the LEU fuel assembly at the beginning of the cycle was also further reduced in comparison to the reference case.

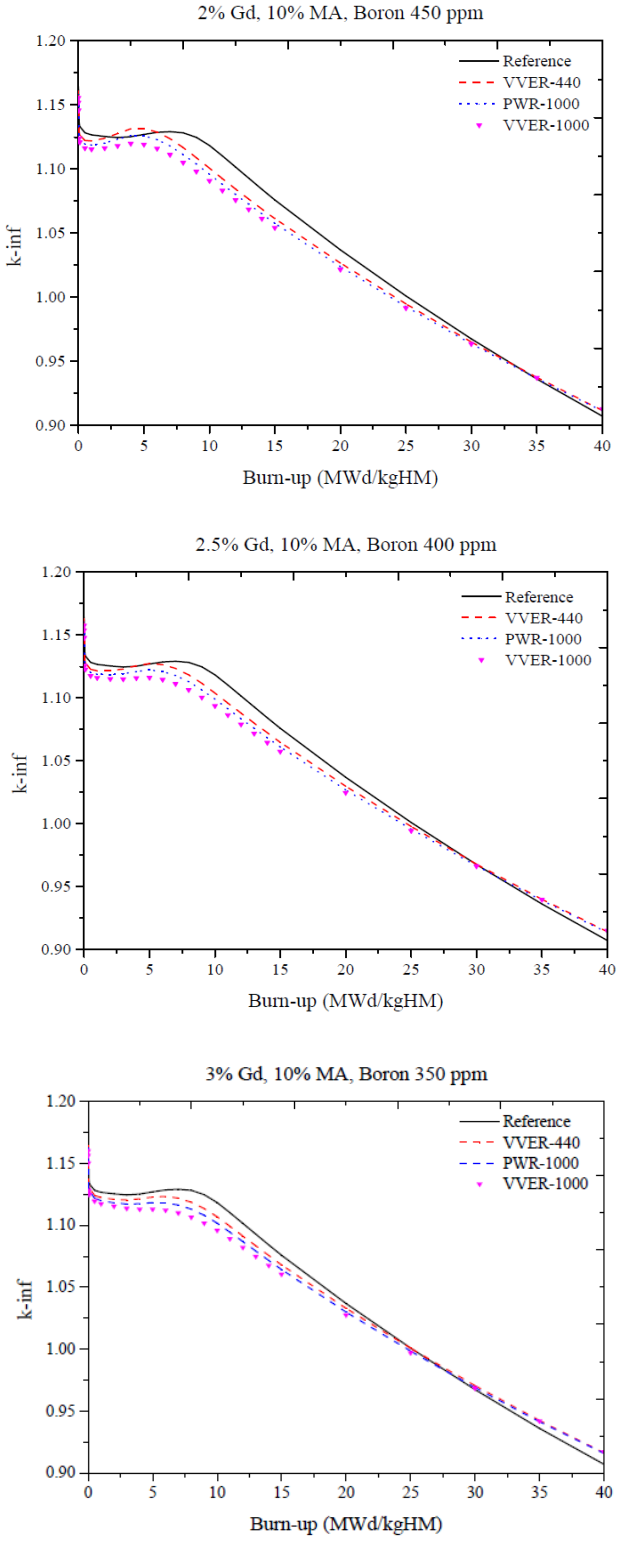

Fig. 3. The k-inf of the LEU fuel assembly versus burnup when loading 10 wt% of MA and reducing GD to 2 wt% (upper), 2.5 wt% (middle) and 3 wt% (lower)

Table II. Transmutation capability in case of homogeneous loading 10 wt% of MA

Isotope	VVER-440 MA vector			PWR-1000 MA vector			VVER-1000 MA vector		
	Initial amount (g)	Mass reduced after 306 days		Initial amount (g)	Mass reduced after 306 days		Initial amount (g)	Mass reduced after 306 days	
		(g)	(%)		(g)	(%)		(g)	(%)
^{237}Np	896.78	140.19	15.63	765.09	118.35	15.47	0.00	—	—
^{241}Am	580.05	223.76	38.58	877.75	313.70	35.74	1536.57	482.05	31.37
^{243}Am	269.52	49.80	18.48	158.24	26.79	16.93	241.75	38.26	15.83
^{244}Cm	81.54	-42.09	-51.62	29.94	-28.17	-94.09	50.18	-39.56	-78.84
^{245}Cm	4.79	-4.94	-103.14	1.65	-3.10	-187.40	4.17	-3.90	-93.42
Total	1832.67	366.73	20.02	1832.67	427.57	23.33	1832.67	473.97	25.86

The results illustrated in Fig. 3 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 up 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 reduced to around 2.5-3.0 wt% and the boron concentration reduced to around 350-400 ppm. As a result, loading 10 wt% of MA 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.

It is found that the case of loading MA vectors from the VVER-440 shows the highest k-inf while that from the VVER-1000 exhibits the lowest k-inf. This also makes the excess reactivity at the beginning of the cycle when loading the MA vector from the VVER-440 spent fuel higher than the two others. However, Fig. 3 indicates that the fuel cycle length is mostly unaffected when loading with different MAs vectors.

The transmutation of MA isotopes is shown in Table II for the cases when loading 10 wt% of MAs and adjusting the gadolinium content to 3 wt% and the boron concentration to 350 ppm. It can be seen that the concentrations of ^{241}Am and ^{243}Am decreased with fuel burnup while those of ^{244}Cm and ^{245}Cm accumulated with fuel burnup. The concentration of ^{237}Np decreased with burnup when loading the VVER-440 and PWR-1000 MA vectors. After 306 days, the ^{237}Np concentration was reduced ~15.63 % when loading the VVER-440 MA vector and ~15.47 % when using PWR-1000 MA vector. The ^{241}Am concentration reduced ~38.58 %, ~35.74 % and ~31.37 % while the ^{243}Am concentration reduced ~18.48 %, ~16.93 % and ~15.83 % in correspondence with loading VVER-440, PWR-1000 and VVER-1000 MA vectors. Meanwhile, those of ^{244}Cm and ^{245}Cm increased ~51.62%, ~94.09 %, ~78.84 % and ~103.14 %, ~187.40 %, ~93.42 % corresponding to VVER-440, PWR-1000 and VVER-1000 MA vectors. 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 total transmutation rate higher than ~20% can be achieved. Besides, it is noticed that in case of loading the VVER-1000 MA vector without ^{237}Np, the transmuted amount of ^{241}Am was much larger compared with the two other cases since the initial loading amount of this isotope was more than two times larger. This explained why the case of loading the VVER-1000 MA vector exhibited the highest total MA transmutatiton mass and efficiency as can be found in Table II. It is also worth noting that more than 90% of the radiotoxidity of MAs from long time storage spent fuel (more than hundred years) come from ^{241}Am (half-life of 432 years). Thus, with the significant amount of ^{241}Am that was transmuted in the VVER-1000 fuel assembly, it could contribute to a significant reduction of radiotoxicity level of the radioactive waste.

B. Coating a thin layer of MAs to the UGD pins

In the case of heterogeneous loading of MAs in the UGD pins of the VVER-1000 LEU fuel assembly, MAs were coated as a thin layer at the outside of the UGD pellets as shown in Fig. 4. The thickness of the cladding was kept unchanged and the outer radius of the UGD region was reduced to accommodate the layer of MAs. For the purpose of MA burning and keeping the fuel cycle length, the MA content of 10 wt% was selected in this investigation. The MA coated layer (see Fig. 4) equivalent to homogeneous loading with 10 wt% of MA 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.

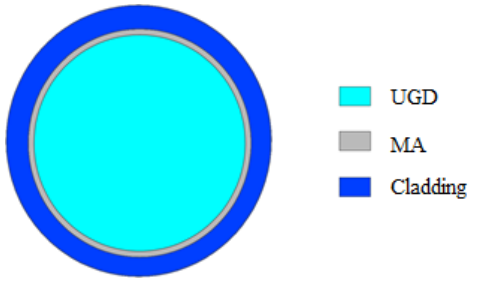

Fig. 4. Coating a thin layer of MA to the UGD pellet

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 Fig. 5 in relation to the reference case. It was found 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 similar to the cases of homogeneous loading as above mentioned. For that reason, the boron concentration was reduced to 400 ppm, 350 ppm, and 300 ppm with respect to the gadolinium content of 2 wt%, 2.5 wt%, and 3 wt%. It is worth noting that the amount of boron concentration reduction in these cases was about 50 ppm larger than the respective ones of homogeneous loading due to the self-shielding effect of MAs. The excess reactivity at the early burnup steps when reducing the gadolinium content to 2 wt%, 2.5 wt%, and 3 wt% was generally lower than the reference case; except that it was slightly higher for the case of the VVER-440 MA vector with the gadolinium content of 2 and 2.5 wt% (Fig. 5). Sooner or later the k-inf in the three cases became smaller than the reference case. However, the cycle length with gadolinium content of 2.5 and 3 wt% was almost the same with the reference case while that with gadolinium content of 2 wt% was somewhat shorter. Consequently,

reducing the gadolinium content to 3 wt% and boron concentration to 300 ppm is recommended when coating with 10 wt% of MA to the UGD pellets. The transmutation of MA isotopes when coating with 10 wt% of MAs and reducing the gadolinium content to 3 wt% and boron concentration to 300 ppm is given in Table III. Comparing Tables III and II, it is shown that the difference in the transmutation rate of MA isotopes between homogeneous and heterogeneous loadings was relatively small. However, the transmutation mass in the case of heterogeneous loading was slightly higher than that with homogeneous loading, in particular for the case of VVER-440 MA vector. Table III also signify that the highest total MA transmutation mass and efficiency was again achieved for the case of loading the VVER-1000 MA vector as compared to the two other cases.

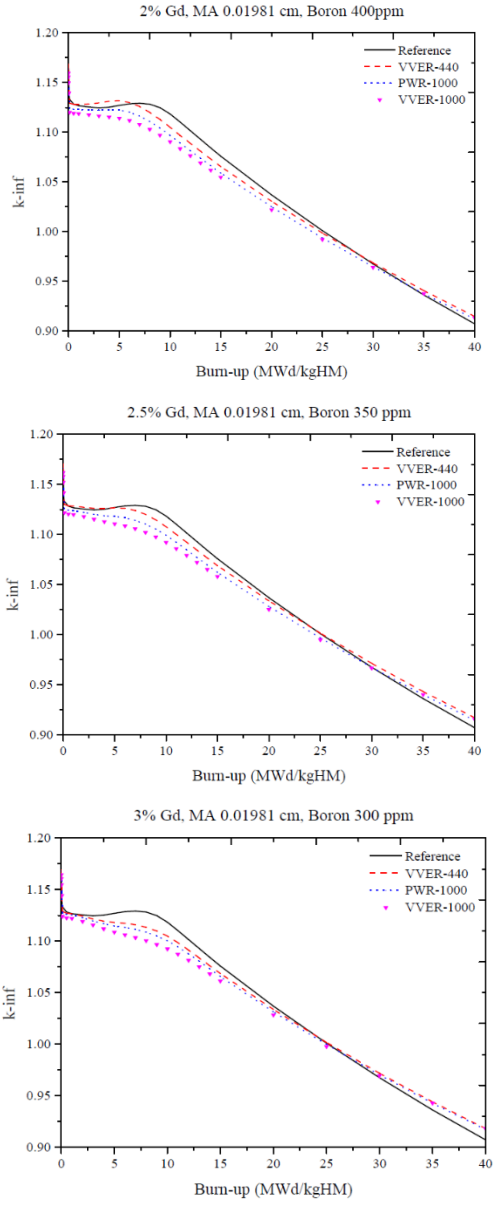

Fig. 5. The k-inf of the LEU fuel assembly versus burnup when coating a layer of MAs to the UGD pins and reducing GD to 2 wt% (upper), 2.5 wt% (middle) and 3 wt% (lower)

Table III. Transmutation capability in case of heterogeneous loading of 10 wt% MA

Isotope	VVER-440 MA vector			PWR-1000 MA vector			VVER-1000 MA vector		
	Initial amount (g)	Mass reduced after 306 days		Initial amount (g)	Mass reduced after 306 days		Initial amount (g)	Mass reduced after 306 days	
		(g)	(%)		(g)	(%)		(g)	(%)
^{237}Np	896.79	150.40	16.77	766.06	118.35	15.45	0.00	__	__
^{241}Am	580.05	238.34	41.09	877.12	317.87	36.24	1536.83	494.08	32.15
^{243}Am	269.51	51.13	18.97	157.98	24.03	15.21	241.56	34.39	14.23
^{244}Cm	81.53	-41.27	-50.62	29.87	-25.87	-86.61	50.12	-36.42	-72.67
^{245}Cm	4.79	-7.81	-142.27	1.65	-3.44	-208.80	4.17	-4.85	-116.46
Total	1832.67	391.79	21.38	1832.67	430.93	23.51	1832.67	484.39	26.43

IV. CONCLUSIONS

In this study, the efficiency of MA transmutation as burnable poison in the VVER-1000 LEU fuel assembly was examined using the SRAC code for the MA homogeneous and heterogeneous loading parterns with different vectors of MAs recycled from the spent fuel of VVER-440, PWR-1000, and VVER-1000 reactors. The gadolinium content and the boron concentration were reduced correspondingly to compensate the negative reactivity insertion by MA loading. The results show that, with 10 wt% of MAs loading, 2.5-3.0 wt% of gadolinium content and 400-350 ppm of boron concentration were recommended for homogeneous mixing MAs in the UGD pins while 3 wt% of gadolinium content and 300 ppm of boron concentration were recommended for heterogeneous loading of MAs in the UGD pins. It was also found that ^{237}Np, ^{241}Am, and ^{243}Am could be significantly transmuted with a transmutation rate as high as ~40% for ^{241}Am. With this transmutation capability, burning MAs in the VVER-1000 fuel assembly could contribute to a significant reduction of radiotoxicity level of the radioactive waste since more than 90% of the radiotoxidity of MAs from long time storage spent fuel (more than hundred years) come from ^{241}Am (half-life of 432 years). Furthermore, the case of loading the VVER-1000 MA vector is highly recommended as it could lead to the highest ^{241}Am transmutation mass as well as the highest total MA transmutation mass and efficiency.

In addition, it was shown that the MAs loading in combination with the reduction in gadolinium and boron concentration could help facilitate the excess reactivity control at the beginning of the fuel cycle without significant effect on the cycle length. Moreover, the MA coating approach could increase slightly the MA burning efficiency when comparing with homogeneous MA mixing model because of the self shielding effect on MAs, especially for the VVER-440 MA vector. Besides, the results indicate that, although loading of different MA vectors slightly affected the fuel cycle length, loading the MA vectors with lower amount of ^{237}Np and higher amount of ^{241}Am could help significantly reduce the excess reactivity at the beginning of the cycle.

Further investigation on transmutation of MAs and radiotoxicity reduction at a full core level and MOX core of the VVER-1000 reactor is being planned.

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