STUDY ON MINOR ACTINIDE TRANSMUTATION IN THE VVER-1000 FUEL ASSEMBLY

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Abstract: Thermal reactors have been considered as interim solution for transmutation of minor actinide 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 actinide 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. 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. Different vectors of minor actinide recycled from the spent fuel of VVER-440, PWR-1000 and VVER-1000 were used in the analysis. The consequent negative reactivity insertion by minor actinide 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 actinide as burnable poison in the VVER-1000 reactor is feasible as minor actinide could partially replace the functions of gadolinium and boric acid for excess reactivity control.

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

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, although the minor actinide isotopes only account for tiny percentage, they 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 minor actinide in the used fuel are indispensable [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 minor actinide 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 minor actinide 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 minor actinide to partially replace the excess reactivity control functions of gadolinium and boric acid.

This study aims to investigate the neutronic feasibility of MA transmutation in a VVER-1000 low enriched uranium (LEU) fuel assembly [17]. The goal is to determine how efficient the MA (Np, Am and Cm) 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 to minimize the cost for fuel fabrication process and respective changes in reactor core design. The SRAC code [18] is used for modeling of the VVER-1000 LEU fuel assembly based on the ENDF/B-VII.0 library. In recent publications, the burnable absorber rods have been suggested as potential locations for loading and burning MAs [15,16] 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 rates. Besides, to estimate the effects of various MAs contents in spent fuel to the VVER-1000 LEU fuel assembly k-inf as well as transmutation efficiency, three MAs vectors from the spent fuel of VVER-440, PWR-1000 and VVER-1000 reactor are used in the analysis [4,15,19].

II. CALCULATION METHOD

The VVER-1000 LEU fuel assembly specified in the OECD VVER-1000 LEU and MOX (mixed oxide) Assembly Computational Benchmark [17] is utilized in the present investigation to examine the feasibility of MA transmutation as burnable poison in the VVER-1000 reactor. The configuration of the VVER-1000 LEU fuel assembly are shown in Fig. 1. The LEU assembly consists of 300 fuel pin cells with 3.7wt% ²³⁵U, 12 UGD pin cells with 3.6wt% ²³⁵U and 4wt% Gd₂O₃, 18 water filled guide tubes for control insertion and one central water filled instrumentation tube. The VVER-1000 LEU fuel assembly is modeled by the SRAC code. The one-sixth of the LEU fuel assembly model in its PIJ module is presented in Fig. 2; the fuel burnup calculations are performed with its BURN-UP module; and the 107 energy groups based on the ENDF/B-VII.0 nuclear data library are used.

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 UO₂ and Gd₂O₃ in the UGD pins; 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 [15,16]. In addition, different vectors of MAs recycled from spent fuel of VVER-440, PWR-1000 and VVER-1000 reactors are used in the study [4,15,19].

The MA vectors consisting of Np, Am and Cm from spent fuel of the VVER-440, PWR-1000 and VVER-1000 [4,15,19] are given in Table 1. The parameters to be investigated are the k-inf 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.

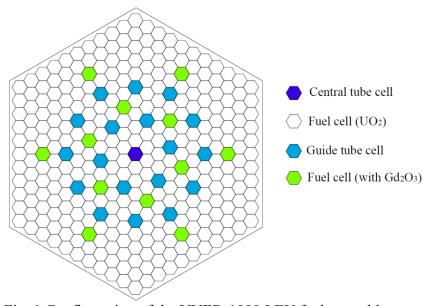


Fig. 1 Configuration of the VVER-1000 LEU fuel assembly

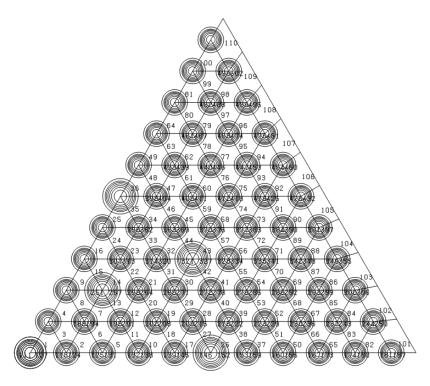


Fig. 2 One-sixth model of the VVER-1000 LEU fuel assembly with SRAC

Table 1 Minor actinide 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 x 10 ⁻⁶	0.03	2.73	0.23	3.59×10^{-6}

III. MA TRANSMUTATION IN THE VVER-1000 FUEL ASSEMBLY

3.1. 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 [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 MAs up to 10 wt% and decreasing the gadolinium content to 2 wt% 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.

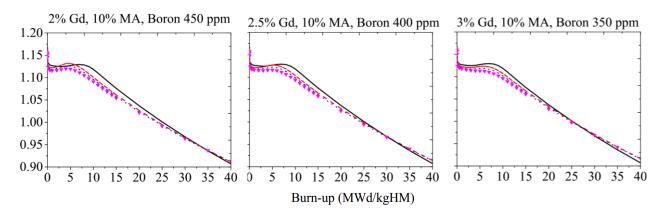


Fig. 3 The k-inf versus burnup when loading 10 wt% of MA and reducing GD to 2 wt%, 2.5 wt% and 3 wt%; Black line – Reference case; Red dash – VVER-440 MA vector; Blue dot – PWR-1000 MA vector; Purple triangle – VVER-1000 MA vector

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 above 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 at the beginning of the cycle was also further reduced in comparison to the reference case.

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 presents the lowest k-inf. This also makes the excess reactivity at the beginning of cycle when loading MA vectors from VVER-440 higher than the two others. However, Fig. 3 shows that the fuel cycle length is mostly unaffected when loading with different MAs vectors.

Isotope	VVER-440 MA vector			PWR-	-1000 MA v	ector	VVER-1000 MA vector		
	Initial amount (g)	Mass reduced after 306 days		Initial amount	Mass reduced after 306 days		Initial amount	Mass reduced after 306 days	
		(g)	(%)	(g)	(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

Table 2 Transmutation rate in case of homogeneous loading 10 wt% of MA

The transmutation of MA isotopes is shown in Table 2 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 ²⁴¹Am and ²⁴³Am decreased with fuel burnup while those of ²⁴⁴Cm and ²⁴⁵Cm accumulated with fuel burnup. The concentration of ²³⁷Np decreased with burnup when loading the VVER-440 and PWR-1000 MA vectors. After 306 days, the ²³⁷Np concentration was reduced ~15.63 % when loading the VVER-440 MA vector and ~15.47 % when using PWR-1000 MA vector. The ²⁴¹Am concentration reduced ~38.58 %, ~35.74 % and ~31.37 % while the ²⁴³Am 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 ²⁴⁴Cm and ²⁴⁵Cm 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.

3.2 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 Fig. 4. 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 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.

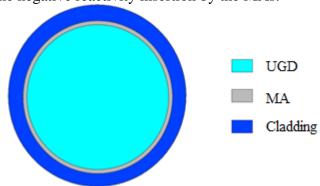


Fig. 4 Coating a thin layer of MA to the UGD pellet

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 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 previously 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 3 wt% was almost equivalent or lower than the reference case; whereas that with 2 and 2.5 wt% was slightly higher (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 3. Comparing Tables 3 and 2, 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..

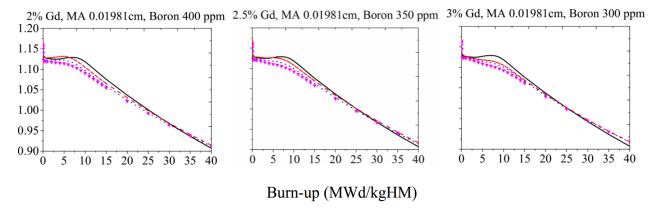


Fig. 5 The k-inf when coating a layer of MAs to the UGD pins and reducing GD to 2 wt%, 2.5 wt% and 3 wt%. Black line – Reference case; Red dash – VVER-440 MA vector; Blue dot – PWR-1000 MA vector; Purple triangle – VVER-1000 MA vector

Table 3 Transmutation rate in case of heterogeneous loading of 10 wt% MA

Isotone	VVER-440 MA vector			PWR-	1000 MA ve	ector	VVER-1000 MA vector		
	Initial	Mass reduced after 306 days		Initial amount	Mass reduced after 306 days		Initial amount	Mass reduced after 306 days	
		(g)	(%)	(g)	(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

The possibility of MA transmutation as burnable poison in the VVER-1000 LEU fuel assembly was examined using the SRAC code. 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% and 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. Besides, the comparison between homogeneous and heterogeneous showed that the coating models could also increase slightly the MA burning efficiency when comparing with homogeneous mixing models because of the self shielding effect on MAs. Consequently, it is highly recommended that transmutation of MAs as burnable poison in the VVER-1000 reactor is feasible taking into account 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 is being planned.

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NGHIÊN CÚU KHẢ NĂNG CHUYỂN HÓA ACTINI HIẾM TRONG BÓ NHIÊN LIỆU CỦA LÒ PHẢN ỨNG VVER-1000

Tóm tắt: Các nghiên cứu trước đây đã chỉ ra rằng lò phản ứng nhiệt được coi là giải pháp chuyển tiếp để chuyển hóa Actini hiếm (MA) từ nhiên liệu đã qua sử dụng. Trong báo cáo này, việc khảo sát các đặc tính vật lý của bó nhiên liệu độ giàu thấp (LEU) của lò phản ứng VVER-1000 trong quá trình chuyển hóa MA được thực hiện. Bó nhiên liệu LEU được mô hình hóa bằng chương trình SRAC. Hai mô hình nạp tải MA trong bó nhiên liệu LEU được thực hiện: trộn đồng nhất và phủ lớp mỏng MA trên các thanh UGD. Các véc-to khác nhau của MA từ nhiên liệu đã qua sử dụng của các lò phản ứng VVER-440, PWR-1000 và VVER-1000 được sử dụng trong phân tích. Độ phản ứng âm do đưa MA vào bó nhiên liệu LEU được bù lại bằng việc giảm hàm lượng gadolini và boron. Hệ số nhân của bó nhiên liệu LEU theo độ sâu cháy, sự chuyển hóa của các đồng vị MA được đưa ra và có so sánh với trường hợp tham khảo. Các kết quả chỉ ra rằng việc chuyển hóa MA trong vai trò chất độc cháy được trong lò phản ứng VVER-1000 là khả dĩ khi MA có thể phần nào thay thế được chức năng của gadolini và axit boric trong việc điều khiển giới hạn độ phản ứng tại đầu chu trình nhiên liêu.

Từ khóa: Bó nhiên liệu LEU của lò phản ứng VVER-1000, chất độc cháy được, chuyển hóa Actini hiếm