

FEASIBILITY OF MINOR ACTINIDE TRANSMUTATION IN THE VVER-1000 FUEL ASSEMBLY

TRAN VINH THANH¹, NGUYEN THI DUNG

Institute for Nuclear Science and Technology – 179 Hoang Quoc Viet str. – Cau Giay Dist. – Hanoi

Email¹: tranvinhthanh.vn@gmail.com

Abstract: The VVER-1000 reactors are nowadays widespread used in Russia and numerous countries. With almost 3000 MW of thermal, VVER-1000 is a candidate to transmute minor actinides (MAs) which are the main contributors to the radiotoxicity and decay heat of nuclear spent fuel (SNF). However, inserting MAs into the reactors has to balance the requirements of reactor long-term service, MAs burning efficiency and safety characteristics. To fulfill those objectives when loading MAs recycled from SNF into VVER-1000, two solutions were suggested in this report: (1) mixing (MA)O₂ homogeneously with UO₂-Gd₂O₃ and (2) coating UO₂-(MA)O₂ in a thin layer outside the UO₂-Gd₂O₃ in Burnable Poison (BP) rods of the VVER-1000 Low Enriched Uranium (LEU) fuel assembly (FA). The calculations were performed by using the Japanese SRAC code system. The results to be presented and discussed are the infinite multiplication factor (*k-inf*) of the FA, and the efficiency of burning MAs in the VVER-1000 LEU FA.

Keywords: VVER-1000 LEU fuel assembly, minor actinide, burnable poison, *k-inf*, SRAC

I. INTRODUCTION

The studies on transmutation of Minor Actinides (MAs) have received much attention in recent years. Although the quantities of MA isotopes (mostly consist of Am, Cm and Np) are small, their effects to neutron source, decay heat and radiotoxicity in spent fuel are substantial [1].

To subside into the harmful effects of MAs in fuel storage, it is necessary to decrease their amount or transmute them into isotopes with shorter half-lives. The previous studies showed that the best way to transmute MAs is using the Accelerator Driven Systems (ADSs). Along with the ADS, the fast neutron reactor can be suggested as an objective to transmute MAs [2-3]. However, the ADSs and fast neutron reactors are the future technologies, using them to transform MA needs certain time from studies to technical applications. Meanwhile, with the huge number of commercial reactors in the world, the light water reactors (LWRs) based on thermal neutron spectra seem to be potential candidates to transmute MAs. Former studies showed that, there were numerous solutions to utilize the thermal reactor for burning MA purpose [4-6]. However, several studies required modifications in the fuel assemblies or core dimensions, those models were difficult to put into mass production due to economic reasons. To overcome that challenge, Hu et al. suggested the direct replacement of MA rods to burnable poison (BP) rods in PWR fuel assemblies. This work only slightly modified the content of fuel and also took the advantages of MA isotopes which have large thermal neutron absorption cross-sections and used them as burnable poisons in the reactor. These results showed that in some sense the MAs played the role of BP and the reactor could reach criticality if it reduced the boron concentration in coolant [7].

The VVER-1000 is nowadays widespread used in Russia and multifarious countries. With almost 3000 MW of thermal, it is a candidate to transmute MAs. In this paper, we studied the effects of MAs on neutronic multiplication factor of the VVER-1000 Low-Enriched-Uranium (LEU) fuel assembly (FA) which is R&D in Russia. The specifications of the LEU FA were gathered from the OECD/NEA benchmark[8].

The MAs were loaded in all BP rods of the LEU assembly. In both two models of this study, the forms of MAs were assumed in oxide powder. Additionally, the MA content was limited not over 10% of fuel mass to satisfy the neutronic characteristics of the VVER-1000 LEU FA. The expected results are the multiplication factor versus burn-up and the transmutation of MAs in the LEU fuel assembly.

II. METHOD AND MODEL

The approach of this study is to find the positions where MA isotopes can be transmuted in the LEU FA by the flux of this reactor while retaining the conventional FA burn-up. Besides, to insert MAs into the reactor, it needs to minimize the fuel design modifications to diminish the additional costs incurred in the manufacturing.

To do the calculation for the VVER-1000 LEU FA, we used the SRAC-2006 code developed by Japan Atomic Energy Agency (JAEA). SRAC is the deterministic neutronic code which integrated 3 transport and 2 diffusion modules for neutron flux calculation. The neutron energy was divided into 107 groups include 74 for fast and 48 for thermal. In addition, the SRAC code provided 12 overlapping groups between fast and thermal groups to help users flexible for choosing cut-off energy [9]. In this study, the LEU FA was modeled by module PIJ which uses the Collision Probability Method (CPM) and Cell Burn-up module which were integrated in SRAC code. The ENDF-7.0 data library with the format in SRAC code was chosen for this calculation. Details of the one sixth LEU FA model in SRAC code were shown in Fig.1.

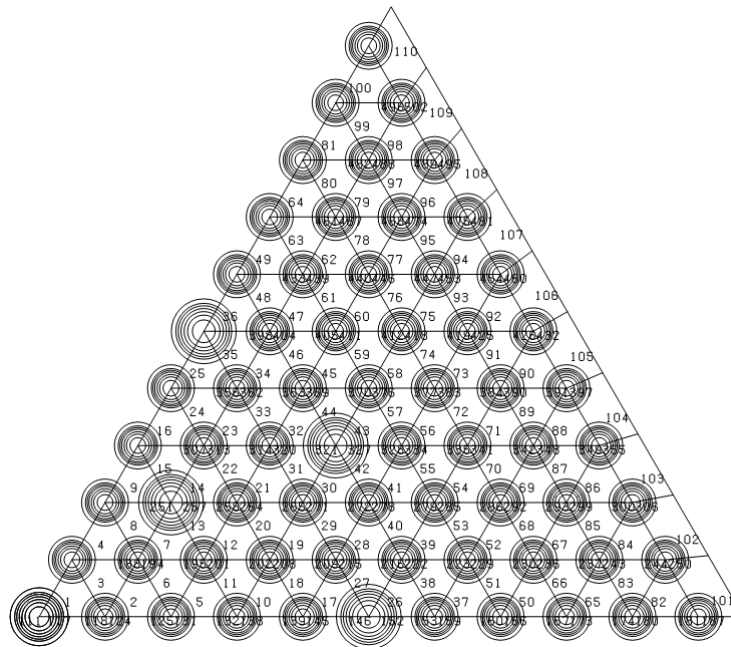


Fig. 1 The one sixth model of VVER LEU FA in SRAC

As mentioned above, the objectives of this study were to find the positions to insert MAs into the LEU FA to burn a quantity of MAs while keeping the dimensions of fuel rods, neutronic characteristics and fuel burn-up of the conventional LEU FA. Thus, in this paper, we proposed 2 models: (1) The MA isotopes were mixed homogeneously with $\text{UO}_2\text{-Gd}_2\text{O}_3$ (UGD) in the BP rods and (2) The UGD in Burnable-Poison (BP) rods were covered by a thin layer of $\text{UO}_2\text{-(MA)O}_2$ (UMA). The purpose of inserting MAs into BP rods in these two models is to annex the neutron absorption ability of BP rods, then reduce the value of $k\text{-inf}$ at first burn-up steps and maintain the fuel burn-up of the FA. The two models of LEU FA when loading MAs were presented in Fig.2 and Fig.3

As mentioned above, the MAs loaded in LEU FA were assumed in the oxide powder for keeping the mechanical properties of material in fuel assembly. Because of the MA vector effects on k -inf and fuel burn-up are negligible, in this study, we used the information of MA vector in the VVER-440 [3]. Those parameters were presented in Table.1.

Table. 1 The MA vector in the VVER-440

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

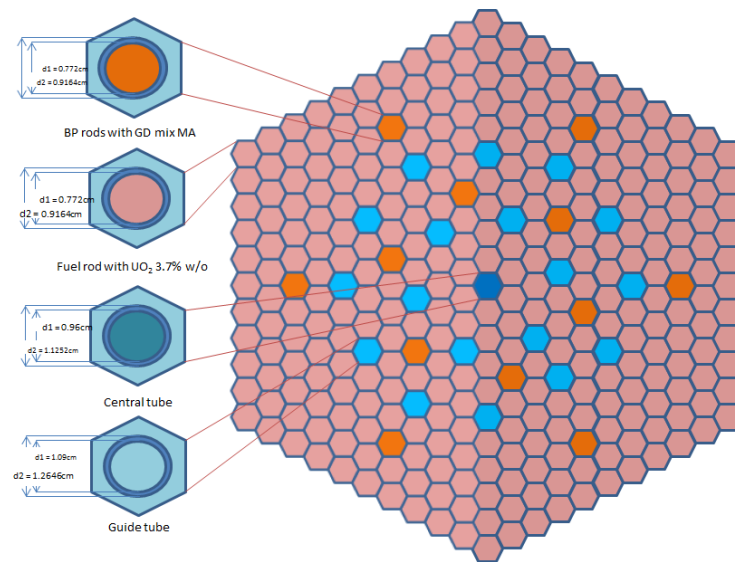


Fig. 2 The homogeneous MA-Gd mixing model

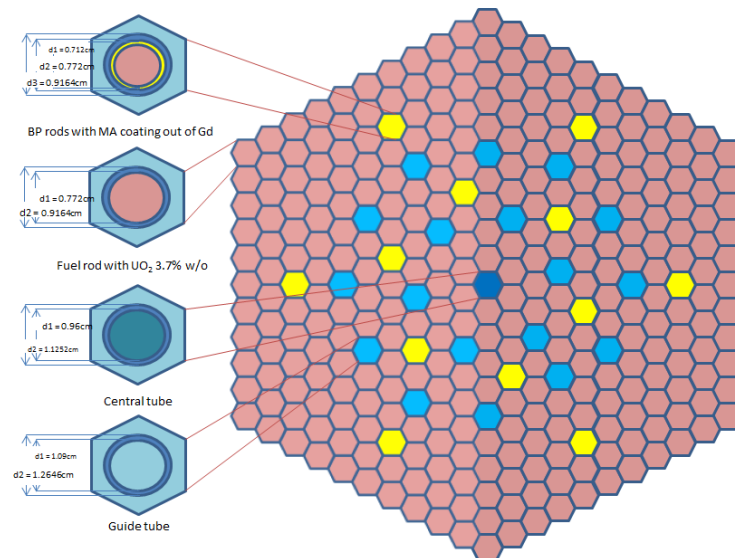


Fig. 3 The MA coating layer model

For convenience, all the calculation procedures of this study were summarized in the flowchart of Fig.4.



Fig. 4 The calculation flow chart

III. RESULTS AND DISCUSSIONS

The results of the FA k_{inf} and transmutation mass of MAs when loading them into the LEU FA were presented in Fig.5 to Fig.9 and in Table.2 to Table.4. In this study, we realized that the cases of mixing UO_2 - Gd_2O_3 -(MA) O_2 could maintain the FA burn-up length to 25 MWd/kgHM as the conventional LEU FA if the concentration of (MA) O_2 was kept lower than 3% fuel mass. For the cases of coating UMA in a thin layer outside the UGD in BP rods, the differences of UMA concentrations and thicknesses did not affect deeply to the k_{inf} . The thicknesses of UMA layer were varied from 0.005 to 0.03 cm and the k_{inf} in those cases were almost similar. Besides, when loading UMA into thin layer, the LEU FA could keep k_{inf} equal to ~ 1.0 at 25 MWd/kgHM.

a. The infinite multiplication factor of LEU fuel assembly

Because inserting MAs to BP rods could increase the thermal neutron absorption cross-section, to maintain the LEU FA burn-up length, in this study we reduced an amount of Gd_2O_3 to balance the neutron absorption. Fig.5 presented the k_{inf} versus burn-up in cases of homogeneous mixing 1-4% MAO_2 with 2% Gd_2O_3 in BP rods.

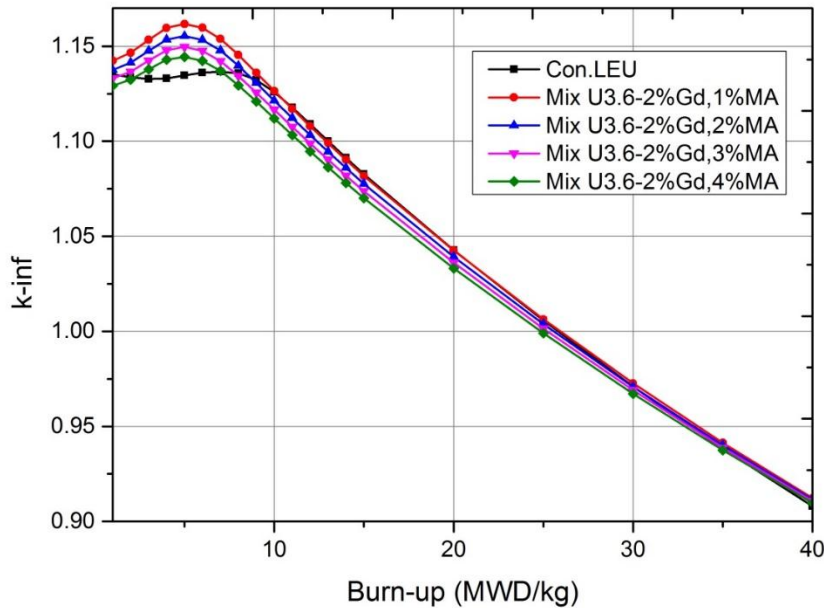


Fig. 5 The k_{inf} versus burn-up in cases of mixing 1-4% MAO_2 with 2% Gd_2O_3

In general, the cases of loading MAs into LEU FA showed that the k_{inf} were lower than that of the conventional LEU from 8 to 35 MWd/kgHM. However after 35 MWd/kgHM, the k_{inf} when loading MA were close to that of conventional LEU FA. The maximum difference between k_{inf} when loading MAs to the conventional k_{inf} was 964 pcm at 8 MWd/kgHM in case of loading 1% MAO_2 while the minimum was 6 pcm at 30 MWd/kgHM in case of loading 2% MAO_2 . It was seen that, from 4 to 8 MWd/kgHM the k_{inf} increased quite rapidly in cases of mixing MA with 2% Gd_2O_3 . When loading 1% MAO_2 , it heightened from 1.1424 to 1.1617 and from 1.1375 to 1.1554 in case of 2% MAO_2 . When the concentration of MAs increased to 3 or 4%, the k_{inf} increased more slowly than cases of loading 1 and 2% MAO_2 . It raised from 1.1292 to 1.1444 when loading 3% MAO_2 and from 1.1331 to 1.1496 in case of 4% MAO_2 .

Naturally, the fuel can avoid damage when the k_{inf} is low at first burn-up steps. Fig.5 showed that when the content of MA up to 3 and 4%, the k_{inf} was kept as low as that of the conventional LEU at 1 MWd/kgHM. The k_{inf} at 1 MWd/kgHM of conventional LEU FA was 1.1310 while in cases of 3% and 4% MAO_2 was 1.1244 and 1.1201, respectively. In addition, when the MAO_2 content was 3%, the k_{inf} was 1.0014 at 25 MWd/kgHM so that keep the fuel burn-up as long as the conventional LEU FA. Meanwhile, the k_{inf} when loading 4% MAO_2 was 0.9902. Thus, in the homogeneous mixing MA-Gd model, to maintain the fuel burn-up length, the content of MAO_2 should not be over 3%.

Fig.6 showed the results of k_{inf} in cases of homogeneous mixing 1-4% MAO_2 with 3% Gd_2O_3 . The curves of k_{inf} when loading MA were in good agreement with that of conventional LEU. In the case of loading 1% MAO_2 , from 9 to 40 MWd/kgHM, the k_{inf} was similar but from 4 to 8 MWd/kgHM, it was higher than that of the conventional LEU. In the cases of loading 2-4% MAO_2 , the values of k_{inf} were lower than in the conventional LEU. However, at 25 MWd/kgHM, while the cases of loading 1-3% MAO_2 provided the k_{inf}

higher than value of 1.0, that of the 4% MAO₂ was 0.9987. Although the *k-inf* when loading 4% MAO₂ was close to 1.0, it was predicted that the content of MAO₂ higher than 4% could reduce the LEU FA burn-up length. The *k-inf* at first burn-up steps in all four cases of mixing (MA)O₂ in BP rods were lower than that of the conventional LEU. In particular, the *k-inf* at 1 MWd/kgHM of the conventional LEU was 1.1351 while those of cases loading 1-4% MAO₂ were 1.1318, 1.1274, 1.1234 and 1.1201, respectively. In cases of loading MA shown in Fig.5 and Fig.6, loading 3% MAO₂ was the most befitting case. That case could satisfy the requirements of the *k-inf* curve characteristic, the burn-up length up to 25 MWd/kgHM and the MAO₂ content loaded up to 3%. Besides, the *k-inf* in that case was always lower than the *k-inf* of the conventional LEU, this warranted the fuel assembly safety in operation.

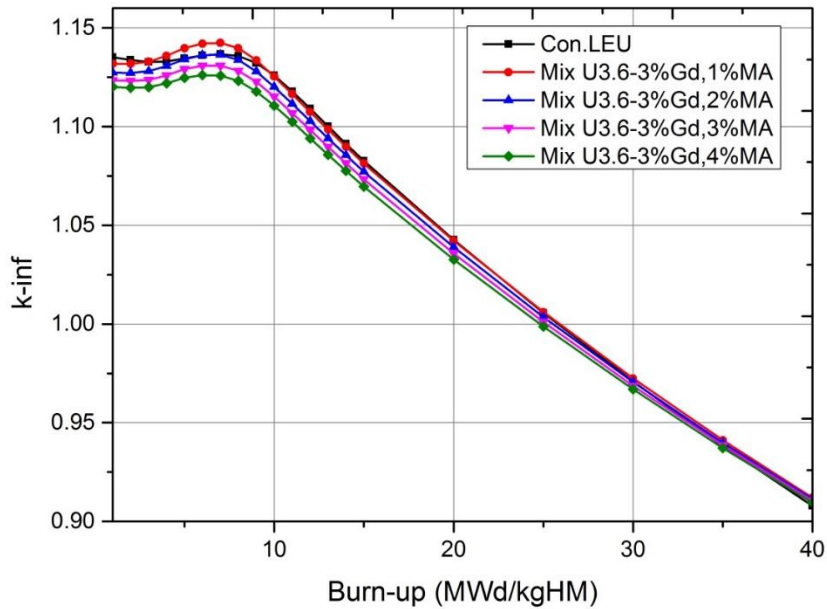


Fig. 6 The *k-inf* versus burn-up in cases of mixing 1-4% MAO₂ with 3% Gd₂O₃

In this study, to estimate the effects of MAs in thin layer on neutronic characteristics of the LEU FA, we did the calculation for cases of coating UMA in layer outside the UGD with the thicknesses varied from 0.001 to 0.05 cm. Besides, the effects of MAs content on *k-inf* were also computed in cases of 2, 4, 6, 8% MAO₂.

Table. 2 The *k-inf* in cases of coating 2, 4, 6, 8% MAO₂ at 0, 10, 20, 40 MWd/kgHM

Burn-up (MWd/kgHM)	3%Gd ₂ O ₃ -2%MAO ₂	3%Gd ₂ O ₃ -4%MAO ₂	3%Gd ₂ O ₃ -6%MAO ₂	3%Gd ₂ O ₃ -8%MAO ₂
	0.001cm	0.001cm	0.001cm	0.001cm
0	1.1788	1.1789	1.1788	1.1788
10	1.1319	1.1319	1.1319	1.1318
20	1.0461	1.0462	1.0461	1.0461
40	0.9128	0.9128	0.9128	0.9128

Table.2 presented the *k-inf* at burn-up steps 0, 10, 20, 40 MWd/kgHM in cases of loading 2, 4, 6, 8% MA in layer with 0.001 cm of thickness, the Gd₂O₃ content in this calculation was 3%. In Table.2, the *k-inf* in all four cases at 0, 10, 20, 40 MWd/kgHM were

almost the same. It showed that when the MA thickness was small, the effect of MA content to k_{inf} was negligible.

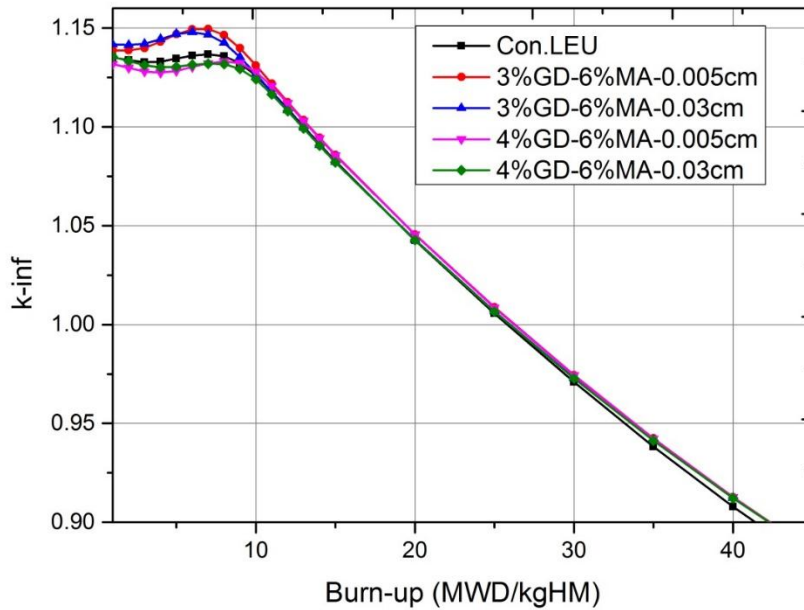


Fig. 7 The k_{inf} when coating UO_2 - MAO_2 with thickness 0.005 and 0.03cm

For the estimation of the MA thickness in BP rods, in Fig.7 the results showed that in range from 0.005 to 0.03 cm, the effects of MA thickness on k_{inf} were small. As shown in Fig.7, when coating 6% UMA outside 3% UGD, the maximum difference of k_{inf} with 0.005 cm and 0.03 cm was 449 pcm, that difference when coating 6% UMA with 4% UGD was 411 pcm. The k_{inf} of all 5 cases in Fig.7 from 9 to 30 MWD/kgHM were almost similar with minuscule differences. However, in case of coating UO_2 - MAO_2 with 3% Gd_2O_3 , the difference of its k_{inf} to that of the conventional LEU from 4 to 8 MWD/kgHM could be up to 1180 pcm at 6 MWD/kgHM. On the other hand, when keeping the content of Gd_2O_3 at 4%, the coating UMA k_{inf} were in good agreement with that of the conventional LEU. The maximum difference of k_{inf} in these case with k_{inf} of the conventional LEU was 468 pcm at 6 MWD/kgHM. The k_{inf} at 25 MWD/kgHM with 0.005 cm and 0.03 cm of MA thickness in case of using 3% Gd_2O_3 were 1.0088 and 1.00673, respectively. These values when using 4% Gd_2O_3 were 1.0086 and 1.0066. Accordingly, in 4 cases presented in Fig.7, the k_{inf} at 25 MWD/kgHM were higher than 1.0. At 1 MWD/kgHM, the k_{inf} was 1.1387 with 0.005 cm and 1.1417 with 0.03 cm of UO_2 - MAO_2 thickness when using 3% Gd_2O_3 . Those values were higher than the conventional LEU k_{inf} (1.1351 at 1 MWD/kgHM). Otherwise, keeping the content of Gd_2O_3 at 4% could give the results of k_{inf} at 1 MWD/kgHM similar to that of conventional LEU. The k_{inf} in case of using 4% Gd_2O_3 was 1.1318 with 0.005 cm and 1.1355 with 0.03 cm of UO_2 - MAO_2 thickness.

As performed, in cases of homogeneous mixing $(MA)O_2$ - Gd_2O_3 , loading 3% MAO_2 with 3% Gd_2O_3 was the most befitting case because it could satisfy the requirements of low k_{inf} at first burn-up step and the fuel burn-up length. In cases of coating UMA in thin layer outside UGD in BP rods, the k_{inf} differences while the thickness of UMA varied from 0.005 to 0.03 cm and the content of UMA changed from 2 to 8% could be neglected. In case of using 3% Gd_2O_3 , the k_{inf} when coating UMA layer were higher than that of conventional LEU in range of 4 to 8 MWD/kgHM. Otherwise, when the content of Gd_2O_3 was 4%, the k_{inf} of UMA coating cases were almost the same to that of the conventional LEU and satisfy the requirements of k_{inf} at first burn-up step and FA burn-up length.

b. The transmutation of MA

Commonly, ^{237}Np has large percentage in MA vector. This isotope also has long decay period ($\sim 10^6$ years). Thus, transmutating the ^{237}Np in reactor plays an important role. In Fig.8, the variations of the ^{237}Np concentration in case of homogeneous mixing MA-Gd were presented.

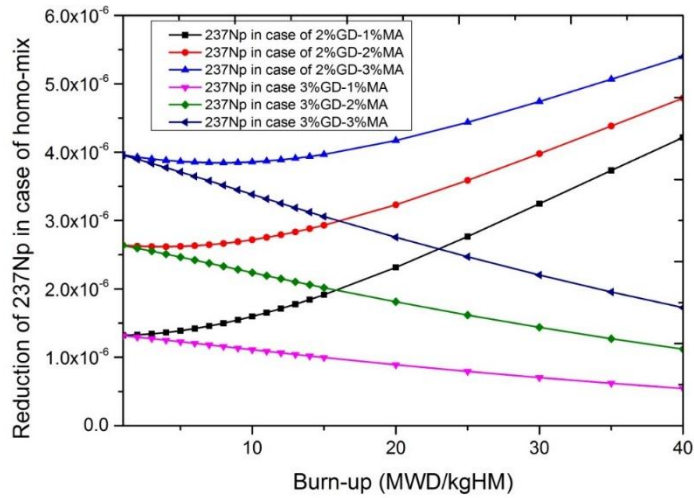


Fig. 8 The concentration of ^{237}Np in cases of mixing 1-3% MA with 2 and 3% Gd₂O₃

It can be seen that, when the content of Gd₂O₃ was 2%, the ^{237}Np concentration increased in burn-up. However, when the content of Gd₂O₃ was 3%, the ^{237}Np concentration reduce versus burn-up. It showed that, to burn the MA effectively, the content of Gd₂O₃ in BP rods should be higher than 3%.

Fig.9 showed the concentration variation versus burn-up of ^{237}Np in case of coating UMA in the layer outside UGD.

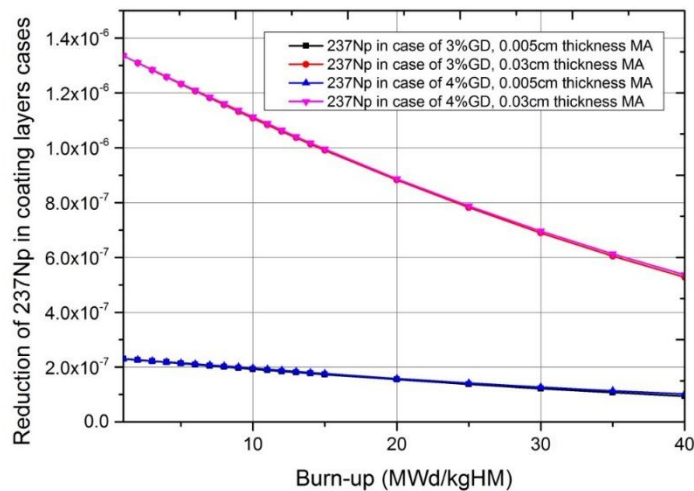


Fig. 9 The k-inf in cases of coating MA

The ^{237}Np concentration in cases of loading MA in thin layer depended mainly on the thickness of UMA. In Fig.9, when the thickness of MA was 0.03 cm, not only the initial MA but also the transmutation rate was higher than that when the thickness was 0.005cm. At 1

MWd/kgHM, the concentration of ^{237}Np with 0.03 cm of thickness more than 6 times when the thickness was 0.005 cm, that value was 5 times at 40 MWd/kgHM.

The masses of MA loaded and discharged after 306 operation days (12 MWd/kgHM) were presented in Table.3 and Table.4, the percentages of MA discharged were also compared with the results on the China AP1000 [10].

Table. 3 The mass and percentage of the MA discharge when mixing 3% MAO₂ with 3% Gd₂O₃

Total discharge of MA in VVER-1000 LEU FA with 3%Gd and 3% MA in BP rods					
Isotope	Initial loading mass (g)	Mass at 306 days (g)	Mass reduced in 306 days (g)	Mass reduced in 306 days (%)	China AP1000 in 300 days
^{237}Np	134.51	108.81	25.70	19.106%	14.399%
^{241}Am	87.00	46.10	40.90	47.011%	42.216%
^{243}Am	40.43	30.47	9.96	24.635%	25.962%
^{244}Cm	12.23	20.76	-8.53	-69.747%	-48.355%
^{245}Cm	0.72	1.83	-1.11	-154.167%	-176.702%

Table. 4 The mass and percentage of the MA discharge when coating 0.03 cm of MA

Total discharge of MA in VVER-1000 LEU FA coating with 4%Gd and 6% MA thickness 0.03 cm in BP rods					
Isotope	Initial loading mass (g)	Mass at 306 days (g)	Mass reduced in 306 days (g)	Mass reduced in 306 days (%)	China AP1000 in 300 days
^{237}Np	45.62	35.66	9.96	21.833%	14.399%
^{241}Am	29.45	13.62	15.82	53.718%	42.216%
^{243}Am	13.67	9.78	3.89	28.457%	25.962%
^{244}Cm	4.13	7.48	-3.35	-81.114%	-48.355%
^{245}Cm	0.24	0.68	-0.44	-183.333%	-176.702%

It was seen that, the trends of MA isotopes transmuted in the VVER-1000 LEU fuel assembly and the China AP-1000 reactor were similar. The differences of MA percentages could be explained by the calculation object in [10] was different to that of this study. In [10], MAs were loaded into every fuel rod of the AP-1000 reactor core while in this study they loaded in BP rods of VVER-1000 FA. Also, the discrepancy of the initial MA vector could make some differences of MA transmutation percentages.

The results in Table.3 and Table.4 showed that the homogeneous mixing MA-Gd model permitted us to load more MA than in the coating layer model. However, the burning efficiencies when loading MA into thin layer were larger than in the homogeneous model. The mass of MA in 0.03 cm thickness in coating layer model was 93.21 g, where the ^{237}Np

mass was 45.62 g. After 306 operation days, the ^{237}Np burnt in LEU fuel assembly was 9.96 g (21.833%). For the homogeneous model, the initial mass of 3% MA in BP rods up to 275.128 g, in there the mass of ^{237}Np was 134.51 g. The mass of ^{237}Np burnt in LEU FA was 25.70 g (19.106%) after 306 days. The results of Table.3 and Table.4 showed that the burning efficiencies of ^{241}Am and ^{243}Am were also considerable. The ^{241}Am was burnt 47.011% in case of homogeneous mixing and 53.718% in coating layer model. For the ^{243}Am , those values were 24.635% and 28.457%, respectively. It was seen that the ^{244}Cm and ^{245}Cm were accumulated in LEU FA, their concentrations increases rapidly in burn-up. In case of homogeneous mixing, the ^{244}Cm and ^{245}Cm masses increased 69.747% and 154.167%, those values in coating layer model were 81.114% and 183.333%. Although the concentration of ^{245}Cm was small, treating this isotope should be considered because of its accumulation and long-live (~8500 years).

III. CONCLUDING REMARKS

In this study, the models of homogeneous mixing $(\text{MA})\text{O}_2\text{-Gd}_2\text{O}_3$ and coating UMA in a thin layer in the BP rods of the VVER-1000 LEU FA were performed. The results presented were the $k\text{-inf}$ versus burn-up and discharged masses of several MA isotopes. It was seen that, in the case of mixing homogeneously $(\text{MA})\text{O}_2\text{-Gd}_2\text{O}_3$, to keep the $k\text{-inf}$ characteristic of the FA containing MA same as the conventional LEU, reducing an amount of Gd_2O_3 in BP rods were suggested.

The results showed that when mixing 3% MAO_2 with 3% Gd_2O_3 , the LEU FA containing MA presented the $k\text{-inf}$ similar to that of the conventional LEU. Besides, the $k\text{-inf}$ at 1 MWd/kgHM when loading MA was smaller than the conventional LEU, so it could guarantee the fuel safety in operation condition. Accordingly, this model could maintain the fuel burn-up as long as that of the conventional LEU.

For the cases of coating UMA in thin layer outside UGD in BP rods, the results showed that the differences of $k\text{-inf}$ when UMA thicknesses varied from 0.005 to 0.03 cm and MA content changing from 2 to 8% were neglected. In order to maintain the fuel burn-up length, the content of Gd_2O_3 in BP rods should be kept at 4%.

For the burning MA requirements, it was seen that the burning efficiencies of ^{237}Np , ^{241}Am and ^{243}Am isotopes were remarkable. However, in the fuel burn-up process, the ^{244}Cm and ^{245}Cm accumulated rapidly was a problem needed to be considered. Although the cases of coating MA layer had higher efficiencies than those in homogeneous mixing model, the MA mass transmuted in the homogeneous mixing model was larger than in coating layer model.

In general, this study proposed two solutions to load MAs in the VVER-1000 LEU FA to transmute apart of MA while keeping the requirements of fuel burn-up length and low $k\text{-inf}$ at first burn-up steps. In the next study, finding the MA loading positions at the FA periphery as well as MAs burning efficiency improvement should be performed.

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

Tóm tắt: Lò phản ứng VVER-1000 hiện nay được sử dụng phổ biến tại Liên Bang Nga và nhiều quốc gia khác. Với công suất nhiệt 3000 MW, đây là một ứng viên tiềm năng để chuyển hóa các đồng vị MA – được coi là thành phần chính đóng góp vào sự nhiễm độc phóng xạ và nhiệt phân rã của nhiên liệu đã qua sử dụng. Tuy nhiên, đưa MA vào trong lò phản ứng cần phải cân bằng được các yêu cầu về thời gian phục vụ, hiệu suất đốt MA và các đặc trưng an toàn. Để thực hiện các mục tiêu đó, trong bài báo này, hai phương pháp nạp tải MA được đề xuất trên đối tượng là bó nhiên liệu độ giàu thấp (LEU) của lò phản ứng VVER-1000, đó là: (1) Trộn lẫn và (2) đặt một lớp mỏng MA bao quanh lớp Gadolina (UO₂-Gd₂O₃) trong các thanh chất độc cháy được (BP). Các tính toán trong báo cáo này được thực hiện bởi bộ chương trình tính toán SRAC của Nhật Bản. Các kết quả được trình bày là hệ số nhân vô hạn (k -inf) và hiệu suất đốt MA trong bó nhiên liệu LEU của lò phản ứng VVER-1000.

Từ khóa: VVER-1000 LEU fuel assembly, minor actinide, burnable poison, k -inf, SRAC