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The Impact of Minor actinides Transmutation in Lead Fast Reactor on Reactor Core Physical Parameters

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Abstract

Long term radiotoxicity produced in the process of nuclear energy utilization has become one of the key problems restricting the safe and rapid development of nuclear energy. Partitioning and transmutation of minor actinides and long-lived fission products (LLFP) are attracting attention at present as an option to reduce the long-term radiological hazard of the high-level nuclear waste. Minor actinides (MAs) are the primary contributors to long term radiotoxicity in spent fuel. As we all know, Fast Reactors (FR) are recognized as one of the most promising method for transmuting MAs, because a large amount of minor actinides can be loaded and the transmutation rate is much higher than that light water reactors. As one of the candidates for the fourth generation advanced nuclear energy system, lead fast neutron reactor (LFR) has great potential for development and can be used for effective transmutation of MAs. However, the addition of MAs in the reactor will have an important impact on reactor physical parameters. Therefore, it is necessary to study the impact of MAs loading in reactor. This work was focused on the impact of MAs transmutation in LFR parameters, such as k_{eff}, β_{eff}, neutron spectrum, et al. The calculations have been carried out with the MCNP5 code. The results show that MAs have significant impact on k_{eff}. The β_{eff} is smaller than without MAs loaded in reactor. The neutron flux and the neutron spectrum do not significantly change with MAs loaded in reactor.

Keywords

Minor actinides, Transmutation, Safety, Lead Fast Reactor.

1. Introduction

The spent fuel unloaded from nuclear power plants has a very long half-life, especially Long-lived High Level Wastes (LHLW), including minor actinides (MAs) and long-lived fission products (LLFP). Among them, those with a long half-life can even reach millions of years [1]. The disposal of spent fuel has become a key factor restricting the development of nuclear power plant. At present, a lot of research work has been done on the glass curing technology of high-level liquid waste containing actinides and the long-term storage of glass solidified blocks [2,3]. However, requiring the final repository to be separated from the biosphere for hundreds of thousands or even millions of years is too long for humans, but so far no country has successfully carried out the final disposal of high-level radioactive waste. There is no clear treatment and disposal plan for a large amount of high-level radioactive waste. Therefore, for nuclear energy to develop in the future, it is necessary to solve

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the major problem of waste disposal. This is a major problem that the international nuclear energy community cannot avoid, and it is also an unsolved worldwide problem [4].

Separation & transmutation technology can separate minor actinides from high-level radioactive waste, and transmutes them into short-lived or stable nuclei in nuclear reactor, and finally eliminating the radioactive hazards [5,6]. At present, Japan, France, the United States, Russia and China have made a lot of efforts to the study of MAs transmutation using nuclear facilities [7-14]. This includes, Accelerator Driven sub-critical system (ADS) [7,8], Fusion-fission hybrid system [9], Pressurized water reactor (PWR), Thermal Reactor [10], Fast Reactor (FR) [11-14], etc. Among of these facilities, ADS probably has more effective transmutation ability than the other reactors, but subcritical reactor technology is not a mature technology. So, the fast reactor is considered to be the most capable for transmutation MAs. The lead-cooled fast reactor (LFR) is a fast neutron energy spectrum reactor cooled by liquid lead or lead bismuth alloy. It can operate safely in high temperature and low pressure environments because of the high melting point and chemical stability of liquid lead. LFRs have great prospects due to the unique safety advantages over other fast reactors. Therefore it is of great significance to study the transmutation characteristics of MAs in lead-cooled fast reactors [15].

Because the delayed neutron fraction of the minor actinides is smaller than that of ^{235}U and ^{239}Pu, and the fission cross-section and capture cross-section of minor actinides are different to ^{235}U and ^{239}Pu, a large amount of the MAs loaded into the reactor core may alter the physics characteristics in several ways and threaten the safety of reactor. The reactor must remain in critical and safe state after loading the MAs in fuel. This indicates that the transmutation of minor actinides in nuclear reactors would ultimately be limited by criticality and safety constraints. After MA loading to the reactor core, the reactor physical parameters mainly including effective neutron multiplication coefficient k_{eff}, effective delayed neutron fraction β_{eff}, void reactivity coefficient and Doppler constant that would be changed. So, it is need to study MAs impact on these parameters in detail. In this paper, the European lead system (ELSY) was chosen as reference reactor (details presented in section 2.), and Monte Carlo method was used for simulation and calculation of the parameters after MAs loading this reactor.

2. Method and calculation model

2.1 Method

MCNP5 code coupled with the ENDF/B-VI library was used for calculation in this work. The MCNP code is the internationally recognized code for analyzing the transport of neutrons, photons and electrons by the Monte Carlo method. MCNP is particularly useful for complex problems that cannot be modeled by computer algorithms that use deterministic methods [16]. It was mainly used to simulate the neutron transport processes and the critical problem in lead-based fast reactor in this work.

2.2 Calculation model

In order to the study the impact of MAs loading in fast reactor, the European lead system (ELSY) was chosen for calculation model, which was developed within the 6th EURATOM Framework Programme (FP6), aimed at investigating the technical and economical feasibility of a 600 MWe lead-cooled reactor[17]. The main purpose was to demonstrate that it is possible to design a competitive and safe fast critical reactor, used also for waste transmutation, by adopting simple engineered technical features. The Lead Fast Reactor (LFR) is also one of the six promising advanced reactors being considered by the GIF (Generation IV International Forum, [18]), because it could represent a significant step in the evolution of nuclear technology.

The ELSY core contains 427 fuel assemblies with PuO2-UO2 Mixed Oxide fuel (MOX). There are 163 in the inner zone, 84 in the intermediate zone and 180 in the outer zone. Three types of fuel were loaded in the reactor core: 86.6Wt% U in MOX fuel loaded in the inner zone, 83.5 Wt % U in MOX fuel loaded in the intermediate zone and 78.8 Wt % U in MOX fuel in the outer zone [19]. The

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components of MOX fuel in the reactor core are listed in Table 1, and the isotopes fraction of Plutonium and Uranium of MOX fuel are listed in Table 2.

Table1 Components of MOX fuel in the reactor core

Core partition	UO_2(%)	PuO_2(%)
Inner zone	86.2	13.8
Middle zone	83.5	16.5
Outer zone	78.8	22.8

Table 2 Isotopes fraction of Plutonium and Uranium of MOX fuel

Plutonium isotope	Fraction (Wt%)	Uranium isotope	Fraction (Wt%)
^{238}Pu	4.0	235U	0.72
^{239}Pu	54.4	238U	99.28
^{240}Pu	22.8		
^{241}Pu	11.8		
^{242}Pu	7.0		

There are twelve positions (six in the intermediate zone and six in the outer zone) for absorber elements devoted to the reactivity compensation and control. The three-zone active part of the core is surrounded by the radial shielding-reflector zone, which is consists of 144 solid lead assemblies. There is a core barrel with the wall thickness of 50 mm and the exterior diameter of 5.6 m surround the shielding-reflector zone [20]. The geometrical structure of the reactor is shown in Fig.1.

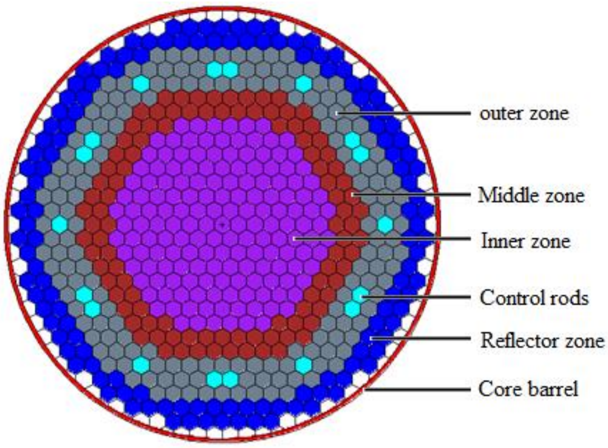

Fig.1 The geometrical structure of ELSY core

3. Results

3.1 The impact of MAs on k_{eff}

In order to analyse the impact of MAs on the reactor k_{eff} , different fraction of ^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm, ^{245}Cm and mix of these MAs were added in reactor used for calculation. The fraction of MAs is 1%, 2%, 3%, 4%, 5% 6%, 7%, 8%, 9%, 10% respectively, and the ratio of each MA nuclide in mix MAs was reference from the depleted fuel of PWR, which is presented in Table 3. The calculation results are shown in Fig.2, the standard deviation of calculation results was less than 0.0002.

Table 3 The ratio of different MA nuclides in the depleted fuel of PWR [21]

MA nuclides	^{237}Np	^{241}Am	^{243}Am	^{244}Cm	^{245}Cm
Ratio	0.56	0.264	0.12	0.0512	0.0028

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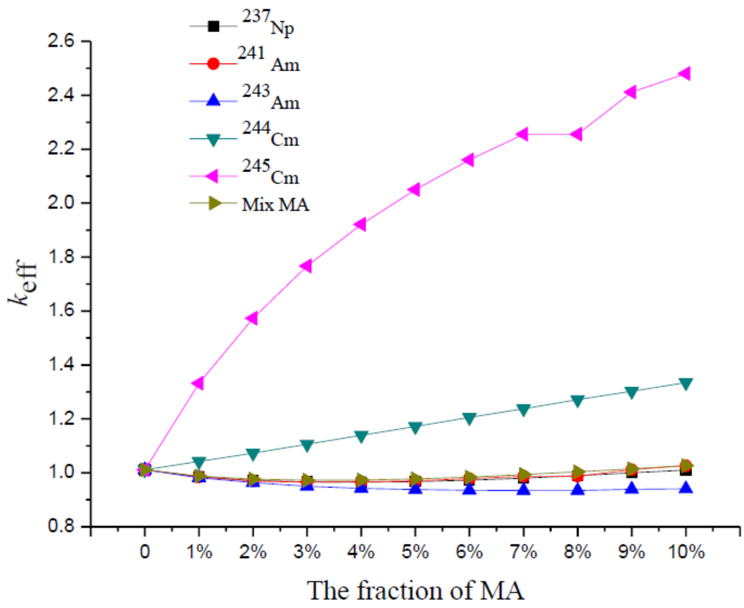

Fig.2. Variation of k_{eff} with different MAs loading fraction

Fig.2 illustrates that each MA nuclide has different impact to the reactor k_{eff}. When ^{237}Np, ^{241}Am and ^{243}Am were added to the fuel, the reactor k_{eff} decreases slightly with the MAs fraction less than 4%, it is because that ^{237}Np, ^{241}Am and ^{243}Am have smaller fission cross-section and larger capture cross-section than ^{235}U and ^{239}Pu (shown as in Table 4), therefore, after adding the MAs to fuel the number of neutrons involved in fission in the core decreases, and the number of neutrons absorbed increases, which leads to the k_{eff} decrease. However, when ^{237}Np, ^{241}Am and ^{243}Am fraction added in the core is more than 4%, the k_{eff} increases slightly with the MAs fraction, it is mainly due to the decrease of total amount of ^{238}U with the increase of MAs fraction, the ratio of capture over fission cross-section of ^{238}U is larger than that of ^{237}Np, ^{241}Am and ^{243}Am. So the number of neutrons absorbed decreases, which leads to the k_{eff} increase.

When ^{245}Cm was added to the fuel, the reactor k_{eff} increases significantly with the MAs fraction, it is because that ^{245}Cm has smaller ratio of capture over fission cross-section than that of ^{235}U and ^{239}Pu. Although the ratio of capture over fission cross-section of ^{244}Cm is greater than that of ^{235}U and ^{239}Pu, the half-life of ^{244}Cm is relatively short (18.1a), and ^{244}Cm has large spontaneous fission cross-section, which can compensate for the neutron loss caused by the large ratio of capture to fission, so the k_{eff} increases with increases of ^{244}Cm.

Table 4 Neutron reaction cross-section of main nuclides in fast reactor (barn)

Isotopes	**^{235}U**	**^{238}U**	**^{239}Pu**	**^{240}Pu**	**^{241}Pu**	**^{237}Np**	**^{241}Am**	**^{243}Am**	**^{244}Cm**	**^{245}Cm**
Fission	2.000	0.044	1.850	0.372	2.630	0.308	0.279	0.242	0.431	2.8
Capture	0.564	0.299	0.514	0.405	0.590	1.58	1.840	1.500	0.673	0.4
Capture/Fission	0.282	6.795	0.278	1.089	0.224	5.130	6.590	6.200	1.462	0.143

3.2 The impact of MAs loading on β_{eff}

Delayed neutrons play an important role in reactor control, and too small delayed neutron fraction is not beneficial to control reactor. In order to study the effect of MAs loading on β_{eff} in detail,TOTNU card was used to calculate the β_{eff}. In a KCODE critical problem, if “TOTNU NO” was written in calculation input file, only prompt neutrons are used for transport calculation, the results were marked as k_{NO}. If “TOTNU” was written in calculation input file, both prompt neutrons and delayed neutrons were used for transport calculation, the results were marked as k_{Yes}. So, the β_{eff} can be calculated by

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$$\beta_{eff} = \frac{k_{Yes} - k_{No}}{k_{Yes} k_{No}} \tag{1}$$

The β_{eff} values were calculated with different fraction (1%, 2%, 3%, 4%, 5%) of MAs, as shown in Fig.3, and the uncertainty of the β_{eff} value was less than 2%. It can be seen that β_{eff} value is decreasing dramatically with the increases of MAs fraction. It is because of that MAs has smaller fraction of delayed neutrons compare with uranium and plutonium. Therefore, it can't load large fraction of MAs in the core, otherwise the safety of the reactor will be threatened. It can be seen that, when the fraction of MAs rises to 4%, the β_{eff} decreases 20% compared with that without MAs. We suggest that the loaded in reactor of MAs fraction should not exceed 5%. Therefore, the fraction of MAs loading in core changes from 1% to 4% in following calculation.

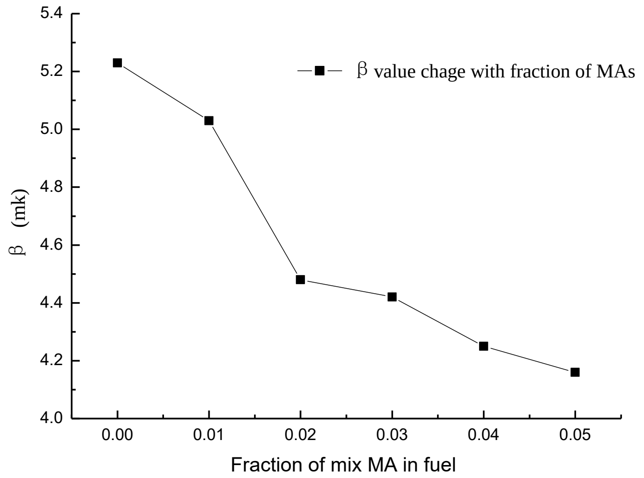

Fig.3. βeff values change with fraction of MAs

3.3 The impact of MAs on neutron flux

In order to study the MAs impact on the neutron flux, the radial and axial direction under different fraction of mix MAs were calculated. The fraction of mix MAs was 1%, 2%, 3% and 4% respectively. Thirteen positions along the radial direction: 0, 20.5, 41, 61.5, 82, 102.5, 123, 143.5, 164, 184.5, 205, 225.5, 246 cm from the core center were chosen for calculation. The normalized neutron flux along the radial direction of different condition was calculated with F5 tally card (the standard deviation was less than 0.0002), shown as in Fig.4. Similarly, thirteen positions along the axial direction: -120,-100, -80, -60, -40, -20, 0, 20, 40, 60, 80, 100, 120cm from the core center were chosen, and the results are shown in Fig.5.

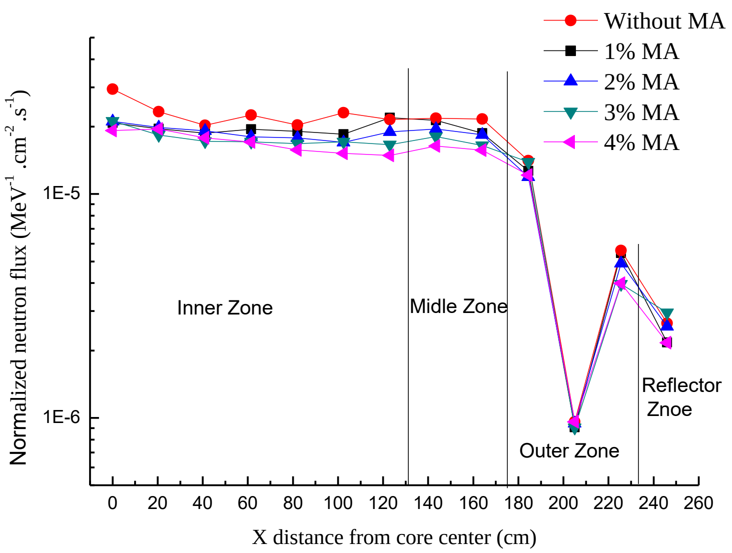

Fig.4. Neutron flux along the radial direction

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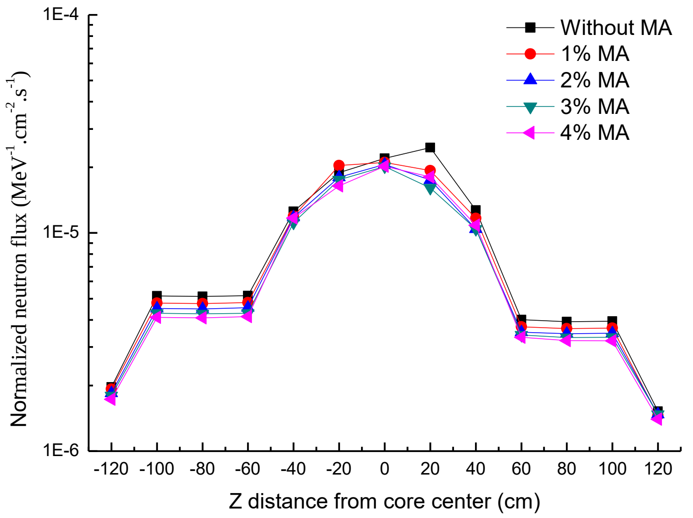

Fig.5. Neutron flux along the axial direction

Fig.4 and Fig.5 indicate that that:1) The neutron flux decreases with the increase of MAs fraction at anywhere, it is due to the large ratio of capture over fission cross-section of MAs; 2) Along the radial direction, the neutron flux decreases slightly at the inner zone and middle zone, and decreases dramatically at the outer zone due to the control rod right in that area, then increases slightly before reflector zone, finally decreases at reflector zone; 3) Along the axial direction, the neutron flux decreases along the center core to both ends, and its shape is similar to that of cosine function which is consistent with the reactor theory.

3.4 The impact of MAs on neutron spectra

Neutron spectrum is critical in transmutation of nuclear waste, the transmutation capability in reactor are mainly determined by the neutron flux and spectrum [22]. The neutron spectra of the ELSY reactor core were calculated by MCNP code with F5 tally card and energy bin card (the standard deviation was less than 0.0002). The results were presented in Fig.6. It shows that the neutron flux decreases with the increases of MAs at low neutron energy region (less than 0.1 MeV), the neutron flux decreases by about 5% with 1% increase of MAs. In fast neutron energy region (exceed 0.1 MeV), the neutron flux almost remains the same.

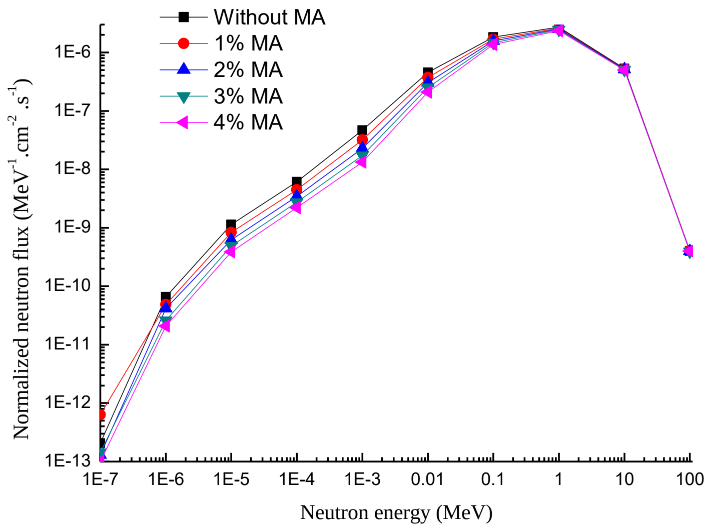

Fig.6. The impact of MAs on neutron spectra

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4. Conclusions

The European lead system (ELSY) core was chosen for calculation model, and the k_{eff}, β_{eff} , neutron flux, neutron spectra were calculated by MCNP5 code under different fraction MAs loading in reactor core. Based the calculation results, the following conclusions can be obtained:

1) After MAs loading in the reactor, it has a significant impact on k_{eff}. The k_{eff} decreases with the increases of MAs fraction.

2) After MAs loading to ELSY core, the β_{eff} decreases corresponding to the increase of MAs fraction. In order to ensure the control safety of the reactor, it is better that the loading fraction of MAs not more than 4%.

3) The neutron flux was no significant change after MAs loading in the reactor whether in radial or axial direction.

4) The calculation results indicate that loading appropriate amount of minor actinides to ELSY core does not disturb the neutron spectra remarkably. In the low energy region (less than 0.01 MeV), the neutron flux decreases, while in the high energy region (more than 1 MeV), the neutron flux has little change.

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