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Research Article

Comparison of the minor actinide transmutation efficiency in models of a fast neutron uranium-thorium fueled reactor*

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Abstract

In terms of nuclear raw materials, the issue of involving thorium in the fuel cycle is hardly very relevant. However, in view of the large-scale nuclear power development, the use of thorium seems to be quite natural and reasonable. The substitution of traditional uranium-plutonium fuel for uranium-thorium fuel in fast neutron reactors will significantly reduce the production of minor actinides, which will make it attractive for the transmutation of long-lived radioactive isotopes of americium, curium and neptunium that have already been and are still being accumulated.

Due to the absence of uranium-233 in nature, the use of thorium in the nuclear power industry requires a closed fuel cycle. At the initial stage of developing the uranium-thorium cycle, it is proposed to use uranium-235 instead of uranium-233 as nuclear fuel.

Studies have been carried out on the transmutation of minor actinides in a fast neutron reactor in which the uranium-thorium cycle is implemented. Several options for the structure of the core of such a reactor have been considered. It has been shown that heterogeneous placement of americium leads to higher rates of its transmutation than homogeneous placement does.

Keywords

Transmutation, minor actinides, spent nuclear fuel, uranium-thorium fuel cycle, radioactivity, biological hazard, spent nuclear fuel storage

Introduction

The main problem of nuclear power engineering today is the nuclear waste disposal. Operating nuclear reactors accumulate so-called minor actinides (MAs), i.e., long-lived radioactive isotopes of americium, curium and neptunium. At present, the MA management all over the world, as a rule, is reduced either to their shipment to storage facilities as part of spent nuclear fuel (SNF) or to their disposal in long-term repositories along with fission products. For the existing and prospective large-scale nuclear power industry, various options of the transmutation fuel cycle are known, which significantly reduce the danger of minor actinides (IAEA-TECDOC-693, Matveyev et al. 1993, Guy et al. 1994, Ganev et al. 2000, Gerasimov and Kiselev 2001, Fazio

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et al. 2013, Kazansky and Romanov 2014, Ivanov et al. 2018, Popov et al. 2018, Korobeinikov et al. 2019).

Up to now, there has been no long-term MA management strategy chosen in any country of the world. Such technologies are being improved and appear to be competitive options for the foreseeable future.

The strategic direction of the nuclear power development in Russia is the nuclear fuel cycle closure based on fast reactors and the reuse of U, Pu and other transuranium elements from SNF of operating reactors.

Russia has accumulated significant experience in reprocessing SNF from thermal reactors. Until now, the reprocessing of SNF from thermal reactors at the RT-1 plant has been accompanied by the release of oxides of uranium, plutonium and neptunium. The long-lived americium radionuclides (^{241}Am and ^{243}Am), along with fission products, remain in high-level waste (HLW). The long-term radiation hazard of the HLW actinide fraction exceeds the radiation hazard of fission products, and its immobilization requires matrices that are especially resistant to leaching.

The separated plutonium, which is supposed to be used for the MOX fuel fabrication, is currently in storage. At the same time, long-term storage of plutonium leads to a deterioration of its isotopic composition, to the loss of its energy value, which is determined by the fissile isotopes (^{239}Pu+^{241}Pu), as a result of β-decay of ^{241}Pu, which leads to the formation of ^{241}Am. The increased content of ^{241}Am in the separated plutonium will complicate its subsequent management (due to the increased heat release and deteriorated radiation characteristics, increased photon and neutron dose rates). For the MOX fuel fabrication, it will be necessary to purify plutonium from americium. As a result, an americium storage facility will appear at the reprocessing plant, and the question will arise: what is to be done with it? Currently, there are two approaches to solving this problem:

1. vitrification of americium together with fission products, followed by final burial in geological formations; and
2. transmutation in reactor systems, which is relevant in terms of reducing its accumulation during the operation of existing thermal neutron reactors.

However, experts still cannot agree on this issue. The main reason for the accumulation of americium is the long-term storage of SNF from thermal reactors.

This paper investigates the possibility of using fast neutron reactors operating in a closed uranium-thorium nuclear cycle for the transmutation of MAs. In particular, the neutronic aspects of solving this problem are studied.

Approaches to minor actinide transmutation and their analysis

Interim SNF storage is much cheaper than reprocessing. However, there is no proof of the safety of long-term SNF storage. In addition, over time, it becomes necessary to build new storage facilities. For example, the SNF storage facility in Zheleznogorsk commissioned in 2011 (if the current rate of irradiated fuel accumulation continues) will be filled by about 2070. The strategic direction for the nuclear power development in Russia is that fast and thermal reactors will operate in pairs, implementing a two-component nuclear power development option (Alekseev et al. 2016). It is obvious that, even before the transition to fast reactors, the recycling of nuclear materials should be arranged. It will be inevitably necessary to deal with fractionation and isolation of MAs, and look for options for their transmutation or combustion in the fuel cycle.

The problem of SNF management is also of international importance. Countries that are starting to develop nuclear power do not want to build reprocessing capacities on their own territories: most of them prefer to give fuel to the supplier of nuclear technologies for reprocessing. Some countries plan to dispose of radioactive waste on their territories while others cannot afford it but they are ready to pay for disposal services. Thus, a complex service is currently in demand, including SNF reprocessing, fresh fuel fabrication and MA disposal.

Thorium fuel cycle

Depending on the price of uranium, as well as the cost of recycling nuclear materials and the costs of implementing the back-end, the thorium fuel cycle may also be commercially attractive in the future. However, until now, this technology has not yet reached a commercial level that would allow free access to the market. Only India is currently focusing on this fuel cycle option (Alekseev and Zaitsev 2014).

Successful full-scale demonstration tests of this reactor technology carried out in the past indicate that there are no insurmountable technical difficulties associated with the use of thorium fuel and the fuel cycle in both existing and evolutionary light water reactors. However, reprocessing and refabrication of uranium-thorium fuel still requires a large amount of research and development work, including the implementation of remote fuel fabrication systems and the provision of sufficient radiation protection and non-proliferation measures as well as industrial infrastructure. So far, there are no research, design and license materials that would allow the use of thorium fuel in existing types of reactors for the short term.

Options for the use of thorium fuel in a closed nuclear fuel cycle based on light water or heavy water reactors, alone or in combination with fast reactors, look more attractive in terms of efficient use of resources. The Generation IV International Forum (GIF) considers molten-salt-cooled reactors with a uranium-thorium fuel cycle as a possible long-term alternative to fast reactors with a uranium-plutonium fuel cycle. However, ^{233}U fully recyclable thorium can be implemented only in the long term, as it still requires a significant amount of research and development work as well as feasibility studies to prove its economic viability.

Fast reactors (FR) can also operate in the uranium-thorium fuel cycle; however, the features of ^{233}U-^{232}Th fuel in the fast neutron spectrum are inferior to the uranium-plutonium fuel cycle in terms of breeding characteristics. Nevertheless, the use of uranium-thorium fuel in FRs can provide a solution to some local problems, such as reducing the void reactivity effect (up to a negative value), reducing the production of transuranium nuclides in the fuel cycle, the production of uranium-233, etc.

Calculation studies of the americium transmutation in a fast U-Th-fueled reactor

For calculation studies on the transmutation of MAs, a modified core of the RBEC reactor (Alekseev et al. 2004) was used, in which the volume fraction of fuel = 0.233, of steel = 0.116, of the coolant = 0.625, and of the gap = 0.028. Uranium-plutonium fuel was substituted for uranium-thorium fuel with the addition of americium. The calculations were carried out using the SERPENT-2 program code and the JEFF-3.1.1 library supplied with this program code (Leppanen 2015). The studies were conducted on three model options of a fast reactor using oxide fuel with the addition of the same amount of americium oxide by weight.

In Option 1, americium oxide was homogeneously mixed into uranium-thorium fuel of the reactor core.

Table 1. Fuel nuclide composition during burnup in Option 1, 10^{-24} nucl/cm^3

Nuclide	Beginning	16 years
U-235.	1.068E–3	3.653E–4
U-232.	0	7.953E–7
U-233.	0	2.145E–4
Th-232	3.244E–3	2.812E–3
Np-237	0	4.181E–5
Pu-238	0	6.361E–4
Pu-239	0	5.613E–5
Pu-242	0	1.808E–4
Am-241	3.123E–3	1.247E–3
Am-243	0	2.046E–3
Cm-242	0	3.105E–3

Table 2. Fuel nuclide composition during burnup in Option 2, 10^{-24} nucl/cm^3

Nuclide	Beginning	16 years
U-235.	0	3.28597E–6
U-238.	3.16144E–3	2.70836E–3
Np-237	0	3.36872E–5
Pu-238	2.10800E–5	6.70935E–4
Pu-239	4.40887E–4	4.11552E–4
Pu-240	3.24056E–4	2.59988E–4
Pu-241	1.45740E–4	3.70718E–5
Pu-242	1.14040E–4	2.81647E–4
Am-241	3.12298E–3	1.18027E–3
Am-243	0	3.65460E–5
Cm-242	0	3.22524E–5

In Option 2, a similar procedure was carried out for uranium-plutonium fuel. The mass of uranium-235 and plutonium in both options was equal. The plutonium vector coincided with the original model of the RBEC reactor and had the form: Pu-238 = 2%, Pu-239 = 42%, Pu-240 = 31%, Pu-241 = 14%, and Pu-242 = 11%. The mass of uranium-238 coincided with that of thorium-232 in Option 1.

Option 3 was heterogeneous with the core divided into two subcores: one with americium oxide and the other with uranium-thorium oxide fuel. The core was divided into two equal volumes, and americium was transferred from the outer core to the inner one, whereas uranium-thorium fuel, on the contrary, to the outer one. In terms of the mass of uranium-235 and thorium-232, Option 3 coincided with Option 1.

Tables 1–4 show the results of calculating the change in the fuel nuclide composition over 16 years of fuel irradiation in a reactor loaded with uranium-thorium fuel with the addition of americium oxide for the three model options described above.

Table 4 for comparison with Option 3 shows the results of the transmutation of americium with account of its isotopic composition. The results show that the transmutation rates of americium-241 in both options are quite close.

Fig. 1 shows the change in the mass of americium over the years of irradiation for the three model options of fast neutron reactor cores. It can be seen from the results that the transmutation of americium has a higher rate for the core with its heterogeneous distribution. This phenomenon can be explained based on the data from Table 5.

Table 3. Fuel nuclide composition during burnup in Option 3, 10^{-24} nucl/cm^3

Nuclide	Beginning	16 years
U-232.	0	4.44E–7.
Np-237	0	4.80E–5
Pu-238	0	1.25E–3
Pu-239	0	1.52E–4
Pu-240	0	1.61E–5
Pu-242	0	3.91E–4
Am-241	6.25E–3	1.72E–3
Am-243	0	5.90E–5
Cm-242	0	6.62E–5
Cm-244	0	1.61E–5

Table 4. Fuel composition change, when the isotopic composition of Am in Option 3 is taken into account

Nuclide	Beginning	16 years
U-232.	0	7.34E–10
Np-237	0	4.24E–5
Pu-238	0	1.13E–3
Pu-239	0	1.40E–4
Pu-240	0	7.04E–5
Pu-242	0	3.54E–4
Am-241	5.60E–3	1.51E–3
Am-243	6.34E–4	2.70E–4
Cm-242	0	5.94E–5
Cm-244	0	1.73E–4
Am-242m	4.40E–6	7.58E–5

Table 5. Spectrum-integrated characteristics of the cores

	U-Th cycle, hom	U-Th cycle, het	U-Pu cycle, hom
Eav	3.28E–1	3.83E–1	3.21E–1
σfis	3.64E–1	4.43E–1	3.53E–1
σcapt	1.09E0	9.84E–1	1.13E0
α	2.99E0	2.22E0	3.19E0
Pfis	2.51E–1	3.10E–1	2.38E–1

Table 5 presents the spectral average characteristics for americium in fast reactors for the considered Am transmutation options. These characteristics include:

- σ_{fis}, σ_{capt}: the spectrum-average fission and capture cross sections;
- P_{fis}: spectrum-average fission probability;
- α: the capture-to-fission cross section ratio;
- E_{av}, MeV: spectrum-average neutron energy.

The heterogeneous variant was averaged over the spectrum of the core with Am. The data in Table 5 show that the average neutron energy in the core with a heterogeneous arrangement of americium is higher than in the other two options. Therefore, the fission cross section is higher than in the two remaining options. The sum of the fission and capture cross sections is also higher for the option with a heterogeneous distribution of americium, which means that the americium transmutation process will go faster. Fig. 2 shows the change in K_{eff} for the calculated systems.

Fig. 3 shows for comparison the plutonium vectors of the considered options of fast neutron reactors using uranium-thorium fuel (Option 1) and uranium-plutonium (Option 2) fuel. It can be seen from the research results that in both options, after 16 years of irradiation, approximately the same amounts of plutonium-238 are formed but, for the option with uranium-thorium fuel, significantly smaller amounts of plutonium-239 and plutonium-240 are produced. Thus, the transmutation of americium in a uranium-thorium reactor results in sufficiently pure plutonium-238, which is a fairly popular material for radioisotope energy sources (RITEGs).

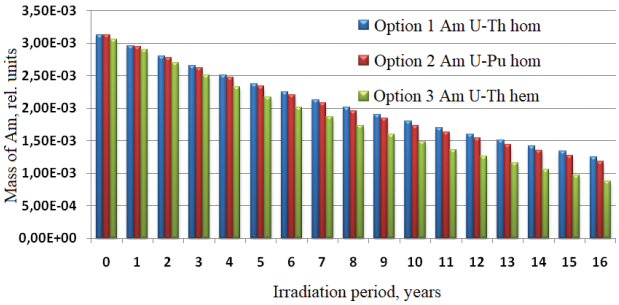

Figure 1. Change in the mass of americium in different model options of transmutation over 16 years.

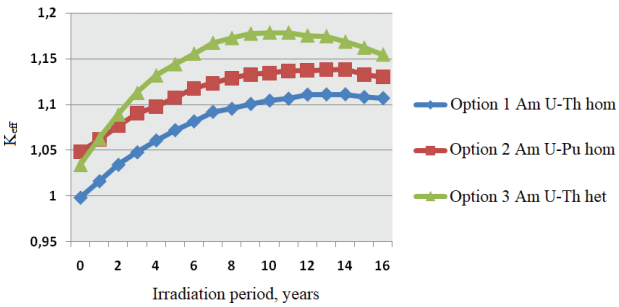

Figure 2. Change in K_{eff} depending on the duration of irradiation for the options of Am-reactors.

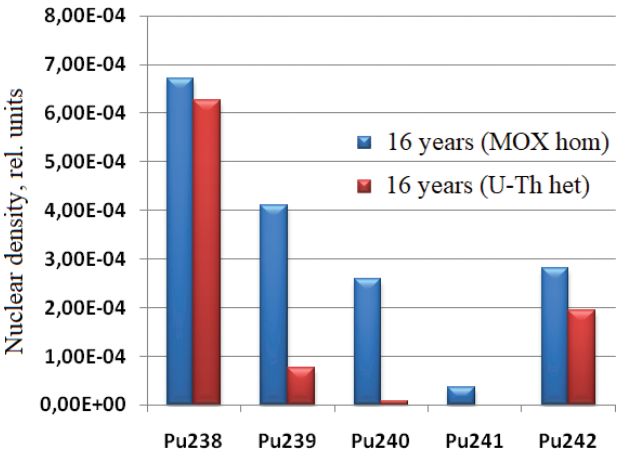

Figure 3. Plutonium nuclide contents in different transmutation options.

Conclusion

The present work describes the studies carried out on the transmutation of americium in fast neutron uranium-thorium fueled reactors. The advantages of implementing this approach to transmutation as compared to the traditional uranium-plutonium approach are quite obvious. Thus, for example, a traditional fast reactor using uranium or MOX fuel for transmutation, in addition to the "external" americium, will additionally produce its "internal" one.

An interesting additional effect of using americium for transmutation in a fast U-Th fueled reactor is the production of sufficiently pure plutonium-238, which is in demand for spacecraft, medical and other purposes.

The problem of developing a real design of such a reactor fueled with uranium-thorium remains open. There is a growing interest in developing reactors implementing the uranium-thorium cycle not only in Russia but also abroad. However, there are no industrial facilities yet.

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