





Research Article

Computational simulation of minor actinide burning in a BN-600 reactor with fuel without uranium and plutonium*

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Abstract

The paper presents the results of studies on the burning of minor actinides (MA) extracted from SNF of thermal reactors in a BN-600 reactor, which uses the complete set of MAs instead of traditional nuclear fuel types: uranium and/or plutonium. The advantages of such approach to MA burning are that long-lived waste is recycled and energy is produced that can be used, e.g., to generate electricity. Besides, where, e.g., a reactor with uranium or MOX fuel is used for transmutation, apart from burning "foreign" minor actinides, it will additionally generate "its own" MAs. The studies have shown that such reactor can be efficient only if based on fast neutrons, which is due to the specific properties of the minor actinide neutron capture and fission cross-sections as compared with traditional fuel nuclides. The calculation results have shown rather a high rate of MA transmutation and burning in a reactor fueled with minor actinides.

Keywords

transmutation, minor actinides, Am, Np, Cm, burning of minor actinides, fast neutron nuclear reactor, neutron spectrum, plutonium, nuclear fuel, SNF, SNF storage, biological hazard, radiotoxicity

Introduction

Disposal of radioactive waste in nuclear power is becoming a growinglysensitiveissue as spent nuclear fuel builds up, the nuclear reactor fleet is enlarged, and NPP units are decommissioned after their design life expires. Specifically challenging in addressing this issue are minor actinides (MA). Two major options are discussed by research community for disposal of nuclear waste, including direct disposal of SNF (an open fuel cycle) with all radioactive elements built up or in the process of build-up, and recycling of long-lived high-level MAs by irradiation in the reactor. The process of transmutation is defined as conversion of MAs and their daughter nuclides to stable or short-lived fission products (Salvatores et al. 1994). Presently, the MA recycling issue has been discussed extensively as part of conferences and in publications on the subject (Salvatores et al. 1994; Kloosterman 1998; Takaki 2000; Bergelson et al. 2005; Youinou and Vasile 2005; OECD NEA 2006; Collins et al. 2007; JENDL-4.0 2010; Hyland and Gihm 2010; Korobeynikov et al. 2019, 2020; Gulevich et al. 2020). Traditional approaches suggest that

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the facility is selected for efficient MA burning. Further, it is required to select the fuel to mix minor actinides into or the positions in these facilities to accommodate irradiation devices with minor actinides. Studies, which used americium-241 and neptunium-237 as fuel, have shown that, in terms of neutronics, such reactor is feasible. The results of these studies are presented in Korobeynikov et al. 2019, 2020. This paper investigates the possibility for the complete set of MAs to be used as fuel rather than individual nuclides.

An important peculiarity of the approach proposed for the MA burning is the fact that MAs as such represent the required source of neutrons, unlike conventional methods when the major sources of neutrons are fuel nuclides such as uranium and plutonium. The efficiency of MA burning in the proposed approach is much higher than offered by conventional approaches.

Note that this paper does not discuss any issues involved in the practical implementation of and the arrangement of operations in such facility. A number of additional studies are required to justify in detail the conceptual approach described. Studies are primarily required for justifying the safety and functionality of FAs with MA fuel.

Properties of minor actinides

MAs are isotopes of americium, curium and neptunium formed as the result of the reactor operation and affecting significantly the SNF and RW handling methods. This is explained by the fact that, despite being produced in small quantities in reactors, MAs contribute greatly to the SNF decay heat power and radiotoxicity in a longer term. Table 1 presents the critical masses and decay heat power values calculated by the authors for nuclides which include as well the minor actinides of interest. Such studies were undertaken also by other authors (Safety Guide No TS-G-1.1 2008; Alajo 2009; IAEA Nuclear Energy Series No. NF-T-4.5 2010). It can be seen from the table that the critical mass of minor actinides is not very large as compared with the nuclides used traditionally as fuel in nuclear reactors. It is therefore possible that minor actinides, as well as the nuclides formed as the result of the neutron capture by these, can be used as fuel in nuclear reactors.

An analysis into the dependence of the capture and fission cross-sections on energy for the nuclides, MAs are composed of, shows (Kosyakin et al. 2021) that

- the fission cross-section for fast neutrons (Am-241 and Np-237) is much greater than the capture cross-section (for other energies, the capture cross-sections are higher than the fission cross-sections);
- a similar dependence of cross-sections exists as well for the Am-243, Cm-242 and Cm-244 nuclides;
- for the Am-242, Am-242, Cm-243 and Cm-245 nuclides, the fission cross-section is higher than the capture cross-section across the region of energies and grows as energy decreases.

Since the Am-241 and Np-237 fission cross-sections are higher than the capture cross-sections only in the "fast" region of energies, and these nuclides account for the most part of the MAs, the reactor with MA fuel can be only a fast neutron reactor. This paper considers the possibility for the **entire set** of minor actinides in the unloaded SNF to be used as fuel, without isolation of americium, neptunium and curium.

Therefore, fast neutron reactors, the fuel in which consists of only minor actinides with no traditional uranium and/or plutonium, will be capable to burn efficiently the MAs accumulated in SNF of thermal neutron reactors.

It is important that most of the MAs in such reactor will be converted to fragments in the process of fission. Another major argument in favor of a reactor with MA fuel is that MA burning does not only eliminate the accumulation of new actinides but also enablesgeneration of electricity. The complication of a traditional nuclear reactor based on uranium or MOX fuel also by the MA burning issue worsens economics and its technical parameters. It is therefore expected that it will be more advantageous to use dedicated reactors the key mission of which is efficient MA burning. Such reactors are naturally expected to be more expensive than traditional reactors. However, they are expected to be capable to burn sufficiently large amounts of MAs, so such dedicated reactors will be few in the nuclear power system.

The results of the studies into the possibility for using Am-241 and Np-237 (Korobeynikov et al. 2019, 2020) as fuel in fast neutron reactors instead of uranium and plutonium have shown a high rate of their burning and rather a high mass of the reactor loading with americium or neptunium-237.

The Table 2 presents the composition of MAs in the unloaded fuel of a VVER reactor after five years of cooling (Szieberth et al. 2012).

Table 1. Critical masses of nuclides in metallic form

Nuclide	<i>T</i> _{1/2} , years	Decay heat power,	Critical mass
Inuclide		W/kg	kg
U-233	1,59E+05	2,81E-01	1,56E+01
U-235	7,04E+08	6,00E-05	4,65E+01
Np-237	2,14E+06	2,10E-02	5,89E+01
Pu-238	8,77E+01	5,60E+02	9,76E+00
Pu-239	2,41E+04	2,00E+00	9,96E+00
Pu-240	6,56E+03	7,00E+00	3,70E+01
Pu-241	1,43E+01	6,40E+00	1,28E+01
Pu-242	3,75E+05	1,20E-01	8,71E+01
Am-241	4,33E+02	1,15E+02	6,18E+01
Am-242m	1,41E+02	4,50E+00	1,26E+01
Am-243	7,37E+03	6,40E+00	1,83E+02
Cm-242	162,8 сут	1,21E+05	1,32E+01
Cm-243	2,91E+01	1,86E+03	6,14E+00
Cm-244	1,81E+01	2,80E+03	2,51E+01
Cm-245	8,50E+03	5,70E+00	1,26E+01
Cm-246	4,76E+03	1,00E+01	8,33E+01

Table 3 presents the results of comparing the critical masses for MA nuclides in metallic and oxide forms. The composition shown in Table 2 was used to calculate the critical mass for the "complete" set of MAs.

 Table 2. Composition of isotopes in unloaded VVER fuel. Five year cooling

Isotope	Mass fraction in SNF	Initial composition for calculations, metallic fue, 10 ²⁴ 1/cm ³	Initial composition for calculations, oxide fuel, 10 ²⁴ 1/cm ³
Np-237	4,95E-1	2,5704E-2	1,1555E-2
Am-241	3,16E-1	1,0786E-2	8,14E-3
Am-242m	1,03E-3	3,501E-5	2,63335E-5
Am-243	1,48E-1	5,01E-3	3,7846E-3
Cm-242	4,13E–9	1,39E-10	1,089E-10
Cm-243	4,36E–4	1,4587E-5	1,14546E-5
Cm-244	3,69E-2	1,22938E-3	9,6592E-4
Cm-245	2,62E-3	8,694E-5	6,8335E-5
Cm-246	4,77E-4	1,576444E-5	1,23963E-5
Cm-247	1,01E-5	3,324163E-7	2,61535E-7
O-16		_	4,91186E-2

 Table 3. Critical masses compared for metallic and oxide MA forms

Nuclide	Oxide, kg	Metal, kg
Np-237	2,82E+02	5,89E+01
Am-241	1,20E+02	6,18E+01
Am-242m	1,58E+01	1,26E+01
Am-243	4,85E+02	1,83E+02
Cm-242	1,84E+01	1,32E+01
Cm-243	7,91E+00	6,14E+00
Cm-244	3,73E+01	2,51E+01
Cm-245	1,47E+01	1,26E+01
Cm-246	1,42E+02	8,33E+01
Cm-247	1,08E+01	8,91E+00
«Вектор» МА	1,93E+02	6,15E+01

Computational studies into ma burning in a bn-600 reactor with fuel of only minor actinides

This section presents the results of computational studies to investigate the efficiency of MA transmutation in a BN-600 reactor model (IAEA-TECDOC-1700 2013), the initial version of which used MOX fuel. For the computational studies into the MA burning efficiency, MOX fuel was substituted by fuel composed of only minor actinides.

Computational model of the BN-600 reactor

Figs 1, 2 show the longitudinal cut and the cross-section of the BN-600 reactor model core respectively. The core consists of zones differing in the U-235 enrichment: a low enrichment zone (center), a moderate enrichment zone (middle), and a high enrichment zone (external). There are steel shielding blocks installed around the core.

Everything in this model that is beyond the core, both axially and radially, is homogenized. The heterogeneous structure has been preserved only for the FAs and the fuel elements within the fuel column. The FA cross-section is shown in Fig. 3.

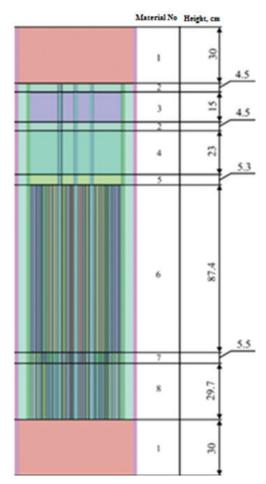


Figure 1. Axial arrangement of materials in the computational model of the BN-600 reactor: 1 - axial reflector; 2 - absorber element end fittings; 3 - absorber elements; 4 - sodium cavity; 5 - fuel element end fittings; <math>6 - core; 7, 8 - lower end shields with depleted uranium dioxide.

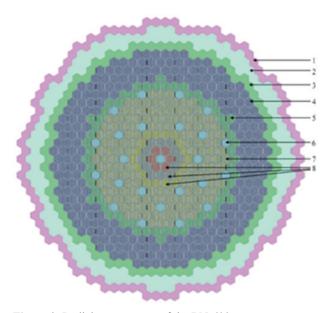


Figure 2. Radial arrangement of the BN-600 reactor core components: 1 – radial reflector; 2, 3 – steel shielding blocks; 4, 5, 7 (8) – FAs with 26%, 21% and 17% U-235 enrichment, respectively; 6 – CPS cluster cell.

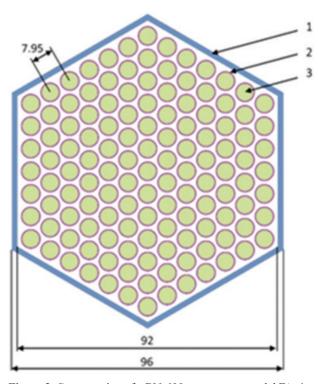


Figure 3. Cross-section of a BN-600 reactor core model FA: 1 – FA jacket; 2 – fuel cladding (wall thickness $\delta = 0.4$ mm); 3 – fuel (diameter d = 6.1 mm).

As fuel, instead of uranium and plutonium, the computational model used the MA composition shown in Table 2. The thermal power was assumed to be equal to 1470 MW.

Results of the computational studies for the MA transmutation in a BN-600 reactor with MA fuel

The computational studies were undertaken for two MA fuel options: in the form of metal and in the form of oxide. SERPENT, a Monte Carlo code, was used for the studies (Leppanen 2015). The SERPENT code took into account the change in the isotopic composition in the course of the reactor operation. In Korobeynikov et al. 2020, the results of comparing the SERPENT code with the MCNP code are presented in the Am burning problem (X-5 Monte Carlo Team 2003). Despite the fact that different libraries were used for the calculations (the JEF-2.2 library (Santamarina et al. 2009) was used for the MCNP calculations, and the standard JEFF-3.1.1 library (Santamarina et al. 2009) was used for the SERPENT calculations), the comparison has shown a good agreement for the results of the calculations using both codes. Fig. 4 presents the comparison of K_{eff} values as a function of irradiation time for the metallic and oxide fuel options.

The K_{eff} value at the irradiation start time grows initially and then drops. This phenomenon is explained by the isotopes produced which prove to be more efficient in terms of contribution to the multiplication factor than the original ones.

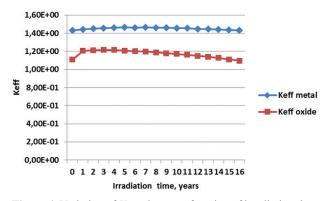


Figure 4. Variation of K_{eff} values as a function of irradiation time for two fuel MA types.

Figs 5, 6 present the change in the mass of the nuclides that contribute the greatest to the MAs in the high enrichment zone in the BN-600 reactor as a function of irradiation time when two MA fuel options (metal and oxide) are used. It can be seen that the trends for the mass change depending on the irradiation time are the same for both fuel forms used in the reactor. The difference in the absolute values is explained by different quantities of MAs loaded into the reactor and by the presence of oxygen in the system with the oxide form that softens the fuel spectrum in the event oxide is used.

Fig. 7 presents the results of the MA change for the total for all reactor zones.

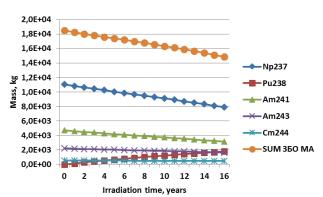


Figure 5. Change in the mass of the nuclides contributing the greatest to the BN-600 MAs (HEZ) as a function of irradiation time for the metallic fuel form.

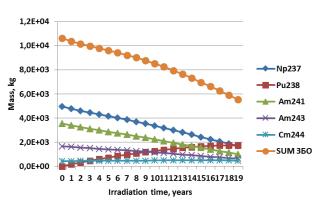


Figure 6. Change in the mass of the nuclides contributing the greatest to the BN-600 MAs (HEZ) as a function of irradiation time for the oxide fuel form.

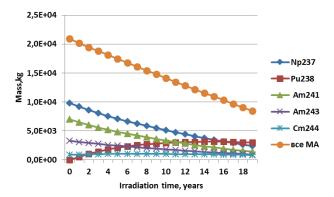


Figure 7. Change in the mass of the nuclides that contribute the greatest to the MAs for all BN-600 zones as a function of irradiation time for the oxide fuel form.

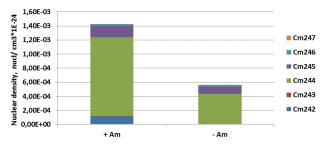


Figure 8. Change in the concentration of Cm nuclides at the end of the irradiation period: +Am – initial option; –Am – americium isolated from the MA composition.

The change in the mass of Cm nuclides depending on the operating time of a nuclear reactor with MA fuel can be traced in Figs 5–7. It can be seen that no burning of curium nuclides is achieved. This is explained by the permanent source for the curium formation from Am nuclides. Especially stable is Cm-244 that contributes the greatest to the MA composition among the curium nuclides. This is rather an important result when different MA transmutation options are used. Fig. 8 shows how the composition of the complete set of Cm nuclides varies for different irradiation scenarios. Two MA irradiation options were considered. Option 1 is +Am – burning of MAs with americium, and option 2 is –Am – isolation of americium from MAs and further burning. It turns out that there is much less curium in the –Am option than in the +Am option.

A question remains what will isolation of Am from the MA composition give? It will be all the same required

afterwards to burn Am separately, and Cm will accumulate again! The solution is to isolate MA from SNF rapidly, before Pu-241, the major source for the Am formation, decays.The faster SNF is processed, the less Am will be contained in MAs.

Conclusions

Neutronic computational studies have been undertaken with respect to burning of minor actinides in a reactor with fuel without uranium and plutonium. Burning of the complete composition of MAs (Am, Np, Cm) isolated from SNF has been investigated.

An important argument in favor of a reactor with minor actinide fuel is that MA burning does not only eliminate the accumulation of new actinides but will also enable generation of electricity. The complication of a nuclear reactor based on uranium or MOX also by the MA burning issue worsens economics and its technical parameters. It is therefore expected that it will be more advantageous to use dedicated reactors the key mission of which will be efficient recycling of MAs. They can be naturally more expensive than traditional ones. It is however expected that it will be possible to burn rather large quantities of MAs in such reactors, so such dedicated reactors will be few in the nuclear power system.

The computational studies were performed using two BN-600 reactor models. One of the models used metallic MA fuel, and the other used oxide MA fuel. It has been shown that MA burning is fairly efficient in both models and with different fuel.

The computational studies have shown that the decay heat from the "complete" set from VVER SNF exceeds that from the same mass of "pure" americium by just 20%.

A major reduction in the mass of Cm nuclides with a high decay power can be achieved by burning MAs with a low Am content. Practical implementation of such approach requires small SNF cooling times prior to the MA isolation since the source of Am-241 is short-lived Pu-241.

The problem of developing a real design of a reactor with fuel based only on minor actinides has not so far been resolved.

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