

Particle emission from irradiated VVER-1200 fuel with Am burnable absorber

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Abstract

For long-term and safe operation of the reactor, nuclear fuel is modified by doping with various homogeneous compounds and heterogeneous inclusions. Such modified fuel has demonstrated satisfactory performance when irradiated at elevated temperatures and high burnup. However, the issues of radiation safety when handling modified fuel remain less studied. Elements of low and medium atomic mass are often targets for (α , n) reactions, so their use as alloying additives, as well as the use of α -emitting additives, can complicate the radiation situation at the stages of the nuclear fuel cycle. In this work, the neutron components of the radiation characteristics of UO_2 with Gd_2O_3 and AmO_2 additives were analyzed. Americium has been investigated as a possible alternative to gadolinium. In fuel containing AmO_2 , the neutron yield is higher compared to Am-free fuel and is formed mainly by (α , n) reactions in AmO_2 in fresh fuel and spontaneous fission of ^{244}Cm nuclides in spent fuel. The research was carried out with the aim of developing procedures and regulations for handling new fuel during its manufacture and after irradiation in the reactor. This work contributes to the study of the neutronic and radiation characteristics of Am-containing fuel, which has the potential for use in modern reactors. Calculations were performed using verified computational codes SOURCES-4C and WIMS-D5B.

Keywords

americium, burnable absorber, neutron yield, radiation characteristics, SOURCES-4C, VVER-1200 reactor, (α , n) reaction

Introduction

In water-water energetic reactors (VVERs), UO_2 with ^{235}U enrichment from 3 to 5% is used as fuel. Uranium-gadolinium fuel in a homogeneous design with axial profiling of fuel rods has received practical application.

For long-term and efficient operation of the reactor, it is proposed to dope the fuel with various homogeneous compounds and heterogeneous inclusions. Studies are being carried out on the properties of the core containing AmO_2 as a burnable absorber (BA) (Shelley and Ovi 2021). Be,

Al, Si, Er, Eu, Dy, Ir, Sm, In, Cm, etc. are considered as alloying additives (Baranov et al. 2011; Kovalishin et al. 2014; Panov et al. 2017; Frybortova 2019a, 2019b; Khrais et al. 2019; Alassaf et al. 2020; Reda et al. 2020; Ovi et al. 2021; Evans et al. 2022; Muzafarov and Savander 2022; Shelley and Ovi 2022). These elements improve the radiation and thermal resistance of the fuel, thermal conductivity, thermophysical and neutronic parameters of the reactor core. For example, when Be is added to fuel, its thermal conductivity improves (Kovalishin et al. 2014). With the introduction of microadditives containing Al and

Si, the grain sizes of densely sintered UO_2 increase and thereby the mechanical strength of the fuel matrix increases (Baranov et al. 2011; Panov et al. 2017). The listed elements have a number of advantages when used as alloying additives, however, some of them are targets for the (α, n) reaction and therefore can significantly complicate the radiation situation at the stages of the nuclear fuel cycle (Vlaskin et al. 1989, 2015; Bedenko et al. 2018a, 2018b; Vlaskin and Khomyakov 2021). This problem is clearly demonstrated by the results of preliminary calculations in the SOURCES-4C program (Wilson et al. 2009) using the “Beam Problems” model, which is a monoenergetic beam of α -particles incident on the target material. See applications of this model in recent studies (Ghal-Eh et al. 2019; Irkimbekov et al. 2021). The energy of α -particles in this case is taken equal to 5.15 MeV, since it corresponds to the weighted average energy of the spectrum of α -particles of spent nuclear fuel (SNF) of the VVER-1200 reactor with a burnup of 60 MW·day/kg(U), cooling for 1 year and a specific activity of $2.58 \cdot 10^{15}$ Bq per fuel assembly (FA).

The yields and spectral distributions of neutrons emitted from light elements in alloying additives for VVER reactor fuel are presented in the graph (Fig. 1) and compared with experimental values (Gibbons and Macklin 1959). For the convenience of the reader, the spectrum of (α, n) neutrons on UO_2 oxygen is also presented (Fig. 1d). In all cases, the integral neutron yields are higher than for oxygen, which is especially noticeable in the case of Be, yield of which is approximately 1000 times higher. In addition, these alloying additives reduce the uranium con-

tent of the fuel pellets (FPs) and complicate the process of their manufacture.

$\text{UO}_2 + 5 \text{ wt.}\% \text{ Gd}_2\text{O}_3$ in homogeneous design has a reduced thermal conductivity value ($\lambda = 3 \text{ W}/(\text{m}\cdot\text{K})$) compared to UO_2 fuel ($\lambda = 4.5 \text{ W}/(\text{m}\cdot\text{K})$) at a temperature of 1000 K (Baranov et al. 2011). The heterogeneous design complicates the fuel production technology, but at the same time the influence of Gd on thermal conductivity is reduced (Tran et al. 2017). However, gadolinium is the strongest absorber; therefore, studies are underway on schemes for its placement in the reactor core in both heterogeneous (Tran et al. 2017) and homogeneous versions (Abu Sodos et al. 2019; Ovi et al. 2021; Muzafarov and Savander 2022).

The search is also underway for alternative BAs, one of which is americium. Fuel with Am is placed in fuel rods without changing the enrichment and does not affect appreciably the power peaking in the FA (Shelley and Ovi 2021), in contrast to uranium-gadolinium fuel (Abu Sodos et al. 2019; Muzafarov and Savander 2022).

The purpose of this work is to determine the neutronic and radiation characteristics of standard and modified VVER-1200 fuel with an increased burnup. To achieve the goal, a neutronic calculation of FAs was carried out, the composition of fresh and spent fuel was calculated, neutron emission yields and their energy spectra were calculated, the equivalent dose rate was assessed, and a comparative analysis was carried out to assess the effect of additives on the radiation background of the fuel. The research was performed using verified computational codes WIMS-D5B (NEA-1507 WIMSD5 2004) and SOURCES-4C (Wilson et al. 2009).

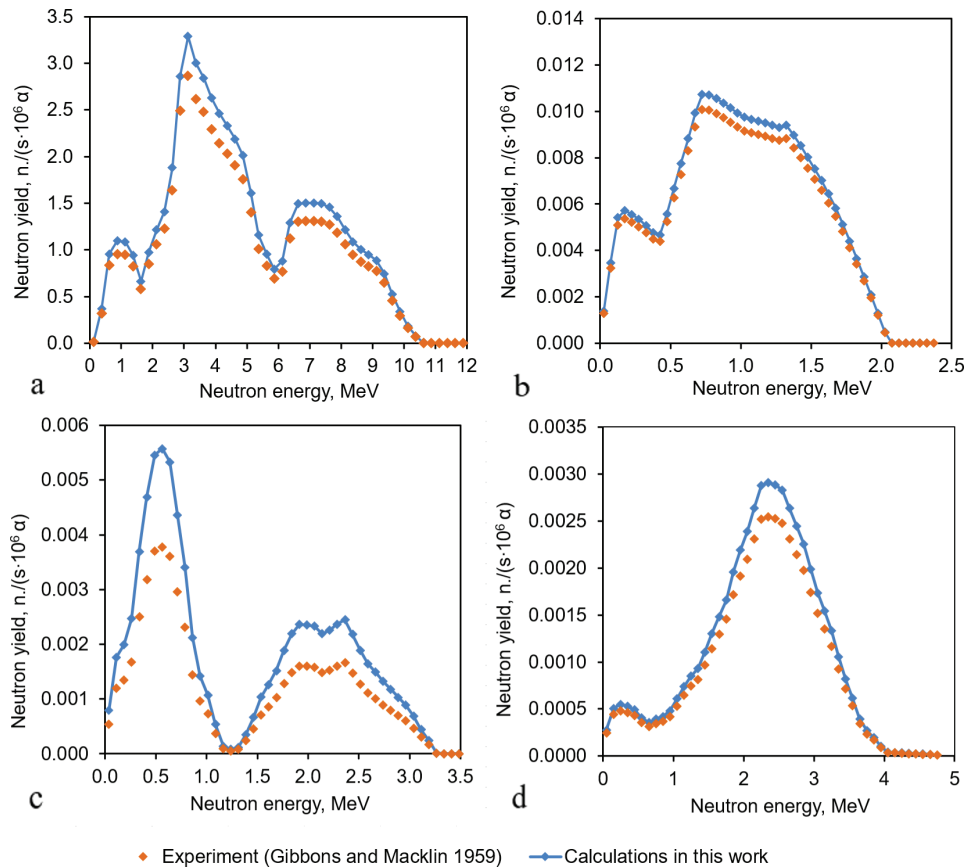


Figure 1. Neutron spectra of elements targeted for the (α, n) reaction: **a.** Beryllium; **b.** Aluminum; **c.** Silicon; **d.** Oxygen.

VVER-1200 fuel assemblies and methods for studying its characteristics

Neutronic model of VVER-1200 fuel assemblies

The calculations were performed using WIMS-D5B code. The program allows the user to carry out neutronic calculations of the core of reactors of various types. A special feature of the program is the ability to take into account fuel burnup during the campaign.

The program uses a universal 69-group library of constants (multi-group cross-sections) prepared on the basis of evaluated neutron data files and containing data for 90 nuclides in accordance with the ENDF/B-VI library (NEA-1507 WIMSD5 2004). The program is capable of solving problems in slab, cylindrical and spherical one-dimensional geometries and in two-dimensional rz-geometry.

The problem of spatial-energy distribution of neutron flux density is calculated in two stages. At the first stage, the real cell is converted into a simplified one, equivalent in area to the real one. Next, the spatial-energy distribution of neutrons in such a cell is calculated as follows:

1. a detailed spectrum is calculated in 69 groups in each of the zones typical for the cell: in the fuel, cladding, coolant and moderator;
2. the cross sections are convolved to a given small-group approximation, in which the detailed spatial distributions of neutrons over the cell are calculated;
3. the resulting solution is modified taking into account leakage;
4. the values are expanded into a 69-group representation, reaction rates are calculated.

The WIMS program implements the collision probability method for solving the neutron transport equation in integral form and the discrete ordinate method, which can only be used in the case of infinite cylinders and plates (NEA-1507 WIMSD5 2004).

The work discusses FAs used on VVER-1200:

1. FA No. 1: 312 fuel rods – UO_2 with 4.95% enrichment;
2. FA No. 2: 312 fuel rods – UO_2 with enrichment 4.95% + 0.2% AmO_2 microcapsules;
3. FA No. 3: 300 fuel rods – UO_2 with 4.95% enrichment, 12 integral fuel burnable absorber (IFBA) rods – UO_2 with enrichment 3.6% + 4.0% Gd_2O_3 . The initial composition of the studied types of FAs is given in Table 1.

The parameters of the FAs are set according to the data used by Shelley and Ovi (2021). The reactor campaign is assumed to be 1468 days. The cells and FAs have a cross-sectional shape of a regular hexagon (Fig. 2a). The WIMS-D5B program uses a FA with equivalent geometry as a calculation model (Fig. 2b). In this case, the cells con-

taining fuel rods, IFBA rods, guide channels, the central tube, as well as the FA itself are circular in plane, since the real hexagonal cells are replaced by equivalent cylindrical ones, with equivalent radii of the FA components shown in Fig. 3 and Table 2. Neutronic calculations in WIMS-D5B are carried out using the first collision probability method. At the outer boundary of the calculation area a reflective condition is imposed.

Table 1. Initial composition of FAs

Nuclide	Concentration, nuclei/($\text{cm}^3 \cdot 10^{24}$)		
	FA No. 1	FA No. 2	FA No. 3
^{234}U	$1.034 \cdot 10^5$	$1.032 \cdot 10^5$	$1.004 \cdot 10^5$
^{235}U	$1.163 \cdot 10^3$	$1.161 \cdot 10^3$	$1.150 \cdot 10^3$
^{236}U	$5.404 \cdot 10^6$	$5.394 \cdot 10^6$	$5.248 \cdot 10^6$
^{238}U	$2.232 \cdot 10^2$	$2.228 \cdot 10^2$	$2.227 \cdot 10^2$
^{241}Am	–	$4.386 \cdot 10^5$	–
^{242}Am	–	$1.669 \cdot 10^6$	–
^{243}Am	–	$8.298 \cdot 10^7$	–
O (nat.)	$4.700 \cdot 10^2$	$4.701 \cdot 10^2$	$4.695 \cdot 10^2$

Table 2. Radii of equivalent geometry, cm

Fuel cell with fuel rod / IFBA rod	Cell with central tube / guide channel
R1 = 0.0600; R2 = 0.3800; R3 = 0.3865; R4 = 0.4550; R5 = 0.6375	R6 = 0.3865; R7 = 0.4550; R8 = 0.6375

Calculation of neutron radiation spectra of VVER-1200 fuel assemblies

The calculations were performed using SOURCES-4C. With this program the intensities and spectra of neutrons from various reactions can be determined. The program code is capable of calculating the radiation intensity of the source used in the (α , n) reaction and neutron spectra in four types of problems:

1. “Homogeneous Mixture Problems” – a homogeneous mixture of an α -radiation source and a target material with a low charge number (Z);
2. “Beam Problems” – a monoenergetic beam of α -particles incident on a plate of target material;
3. “Interface Problems” – an alpha emitter plate located close to a plate of low- Z material;
4. “Three-Region Interface Problems” – a thin plate of low- Z material located between the alpha emitter and the target material.

To output neutron spectra of fuel, the “Homogeneous Mixture Problems” model is selected in SOURCES-4C. The model allows calculations of α -particle transport, neutron yield and spectrum for a homogeneous mixture of α -emitters and light elements. Here, materials subject to α -decay and spontaneous fission are closely mixed with the low- Z target material. Three types of neutrons are considered: spontaneous fission neutrons, delayed neutrons, and neutrons emitted as a result of (α , n) reactions. Neutron source intensity is the number of neutrons produced per second per unit volume.

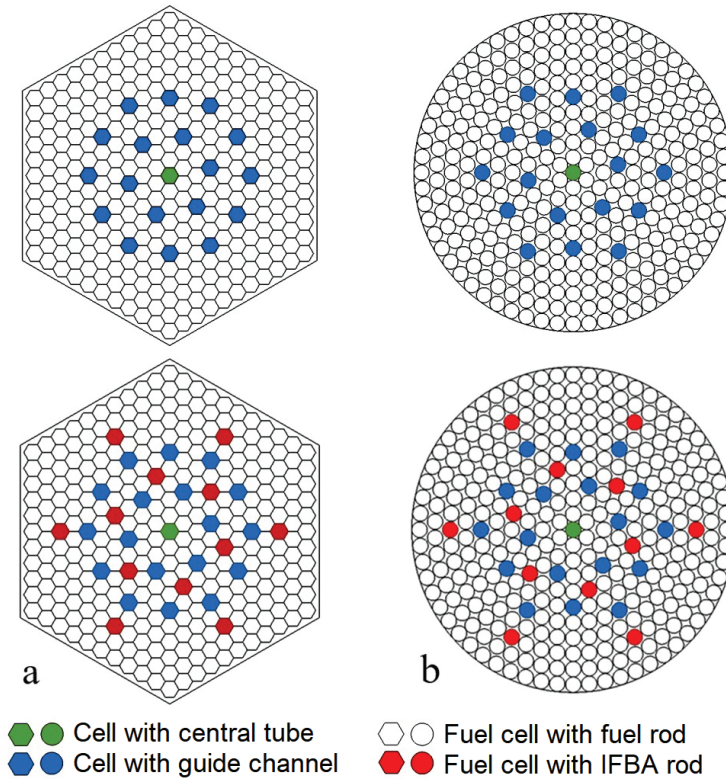


Figure 2. Calculation model of the VVER-1200 FA: **a.** Real geometry of the FA; **b.** Equivalent geometry of the FA.

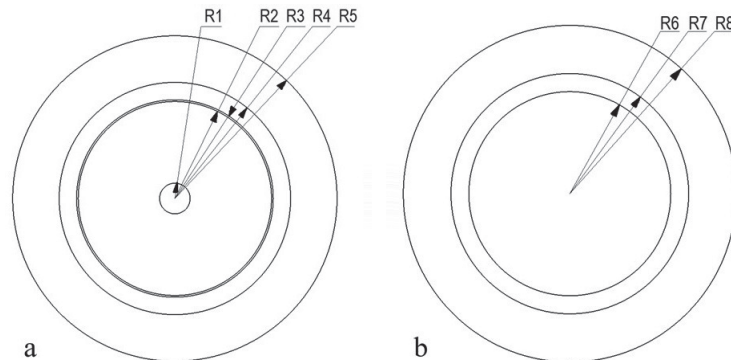


Figure 3. Equivalent geometry: **a.** Fuel cell with fuel rod / IFBA rod; **b.** Cell with a central tube/guide channel (1 – central hole of the FP; 2 – FP; 3 – gap; 4, 7 – shell; 5, 6, 8 – moderator-coolant).

The model assumes that the size of the target is much smaller than the range of α -particles, so that all α -particles stop inside a mixture of target nuclei and α -emitters. The neutron spectra of the (α, n) reaction are determined under the assumption of an isotropic angular distribution of neutrons in the center of mass system (Wilson et al. 2009). Sources of α -radiation are heavy isotopes in the fuel composition (Fig. 4). The target material is a mixture of ^{17}O and ^{18}O – isotopes targeted for the (α, n) reaction. In all cases, the range of neutron energies considered is designated from 0 to 12 MeV and is divided into 48 equivalent monoenergetic groups.

Calculation of radiation characteristics of VVER-1200 fuel assemblies

When calculating the neutron component of the equivalent dose rate, beta and gamma emissions formed in decay pro-

cesses and $(\alpha, n\gamma)$ reactions are not taken into account. The yields and spectral distributions of neutrons $Y(E)$ [$\text{n} \cdot \text{s}^{-1}$], obtained with SOURCES-4C, are used to calculate the equivalent dose rate using the following algorithm:

1. approximation of the neutron radiation spectra using the Watt spectrum for spontaneous fission neutrons, using a 9th degree polynomial for (α, n) neutrons and a normal or lognormal distribution;
2. use of the point geometry approximation: neutron flux density value [$\text{n} \cdot (\text{cm}^2 \cdot \text{s})^{-1}$] inside the FP is equal to that on its surface and is calculated by dividing $Y(E)$ by the surface area of the FP [cm^2];
3. calculation of equivalent dose rate [$\text{Sv} \cdot \text{h}^{-1}$] using conversion coefficients [$\text{pSv} \cdot \text{cm}^2$] depending on the energy of neutrons incident in isotropic geometry (ICRP 2010);
4. normalization of the obtained values per one FA or FP.

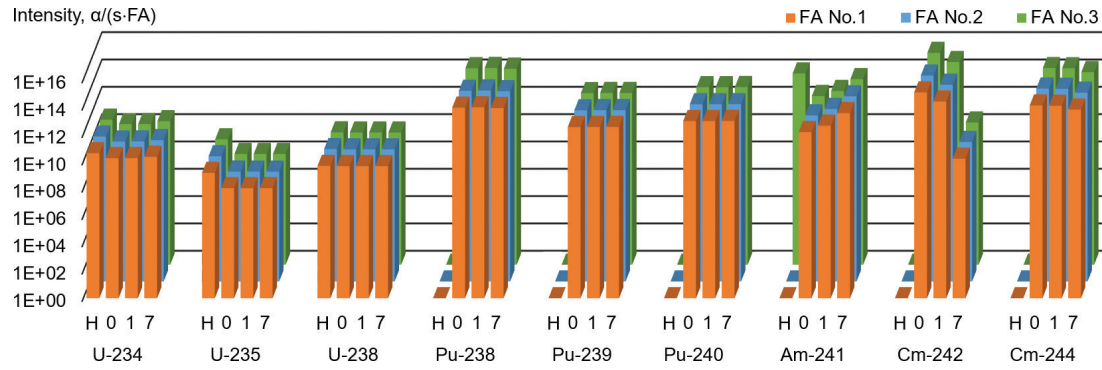


Figure 4. Intensity of α -radiation per fuel subassembly (H – beginning of the campaign; 0 – End of the campaign; 1, 3, 7 – SNF cooling for 1, 3, 7 years).

Results and discussion

Neutronic characteristics of VVER-1200 fuel assemblies

As a result of neutronic calculations, the dependences of k_{inf} (Fig. 5) on time (t) and power generation (z) were obtained for the three FAs under study.

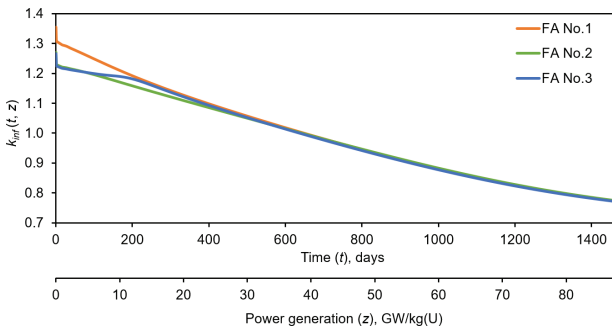


Figure 5. Comparison of $k_{inf}(t, z)$ for fuels with different compositions.

In the case of FA No. 3, rapid burnout of Gd releases reactivity and leads to an increase in k_{inf} . The run-out of reactivity is observed in the range of 100–200 days (Fig. 5). This is typical for FAs with uranium-gadolinium fuel in IFBA rods and is comparable with the results of a number of studies (Tran et al. 2017; Abu Sondas et al. 2019; Ovi et al. 2021; Muzafarov and Savander 2022; Shelley et al. 2022).

Am in FA No. 2 is distributed more uniformly than Gd in FA No. 3, and its macroscopic neutron absorption cross section is lower, so the run-out is practically not noticeable, and the change in $k_{inf}(t, z)$ for the same campaign duration is close to linear (Fig. 5). This behavior of $k_{inf}(t, z)$ was also observed by Shelley and Ovi (2021). The observed decrease in the initial reactivity margin in comparison with FA No. 1 allows us to conclude that the heterogeneous arrangement of AmO₂ microcapsules can be used to compensate for excess reactivity.

Radiation characteristics of VVER-1200 fuel assemblies

Figures 6–8 show the neutron radiation spectra for three types of FAs.

The main source of neutron radiation in SNF is ²⁴⁴Cm nuclei undergoing spontaneous fission. The contribution

of the (α, n) reaction to the spectrum of neutron radiation from SNF is significant only at the end of the campaign, since already after a year of SNF cooling the majority of ²⁴²Cm nuclei, which make the main contribution to the intensity of α -radiation, decay (Fig. 4). In a study conducted by Sharmin et al. (2023), very short-lived nuclides, including ²⁴²Cm, make a significant contribution to the overall radiotoxicity only at the beginning of SNF cooling, and medium-lived nuclides, including ²⁴⁴Cm, make a larger contribution as cooling progresses.

Fresh fuel does not contain curium, so the neutron yield from UO₂ is low due to the long half-life and low probability of spontaneous fission of ²³⁸U in comparison with the ²⁴⁴Cm, as presented in Table 3.

Table 3. Properties of spontaneously fissile nuclides (Shultis and Faw 2002)

Nuclide	Half-life, years	Probability of spontaneous fission per decay, %	Neutrons per fission
²³⁸ U	$4.47 \cdot 10^9$	$5.4 \cdot 10^{-5}$	2.07
²⁴⁴ Cm	18.11	$1.3 \cdot 10^{-4}$	2.77

With increasing fuel enrichment, the integral neutron yield decreases. This is due to the fact that at a lower concentration of ²³⁸U, less ²⁴⁰Pu and, accordingly, less ²⁴⁴Cm are formed through successive nuclear reactions. Therefore, the neutron yield for FA No. 3 is slightly higher (Fig. 9) due to the presence of Gd₂O₃, which requires a reduction in fuel enrichment.

When AmO₂ is introduced, the neutron yield of fresh fuel increases significantly (Fig. 7b) due to the (α, n) reaction, which occurs because ²⁴¹Am is a relatively strong α -emitter (Fig. 4). The contribution of the (α, n) reaction to the neutron radiation of the fuel is presented in Table 4.

During reactor operation, ²⁴¹Am in FA No. 2 burns out, absorbing neutrons. In this case, through successive nuclear reactions, curium isotopes are formed, including ²⁴⁴Cm, which leads to a higher neutron yield of SNF and, accordingly, a higher dose rate compared to FAs No. 1 and No. 3 (Fig. 9). Since the neutron radiation spectra have the same shape during the 7-year cooling period (Fig. 6, 7, 8), the conversion coefficients remain the same, therefore the dependences of the neutron yields and the equivalent dose rate of neutron radiation on the cooling time are similar (Fig. 9). The calculated neutron

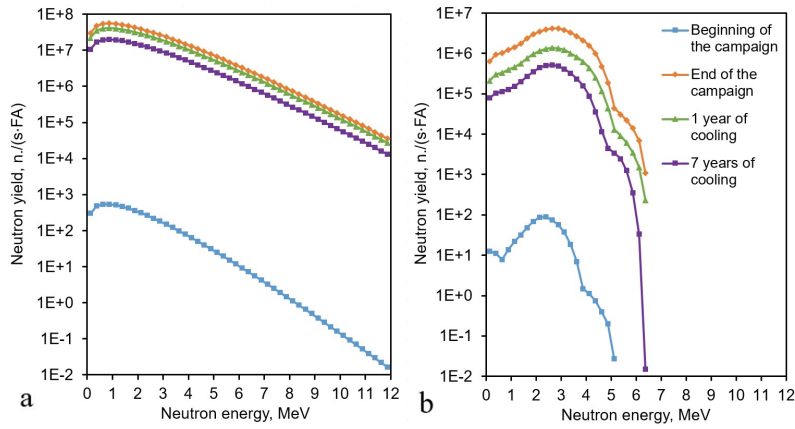


Figure 6. Neutron radiation spectra of FA No. 1: **a.** spectrum of spontaneous fission neutrons; **b.** spectrum of neutrons from (α, n) reaction.

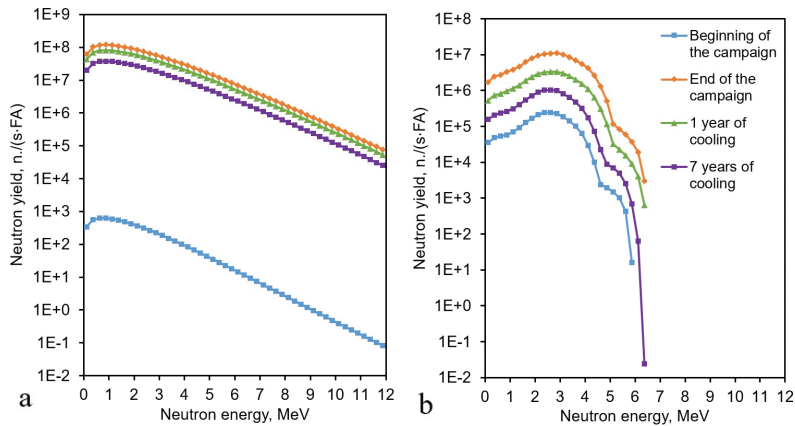


Figure 7. Neutron radiation spectra of FA No. 2: **a.** spectrum of spontaneous fission neutrons; **b.** spectrum of neutrons from (α, n) reaction.

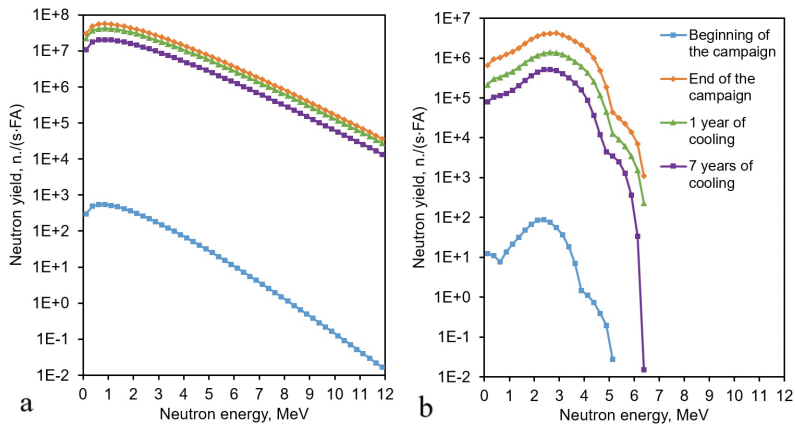


Figure 8. Neutron radiation spectra of FA No. 3: **a.** spectrum of spontaneous fission neutrons; **b.** spectrum of neutrons from (α, n) reaction.

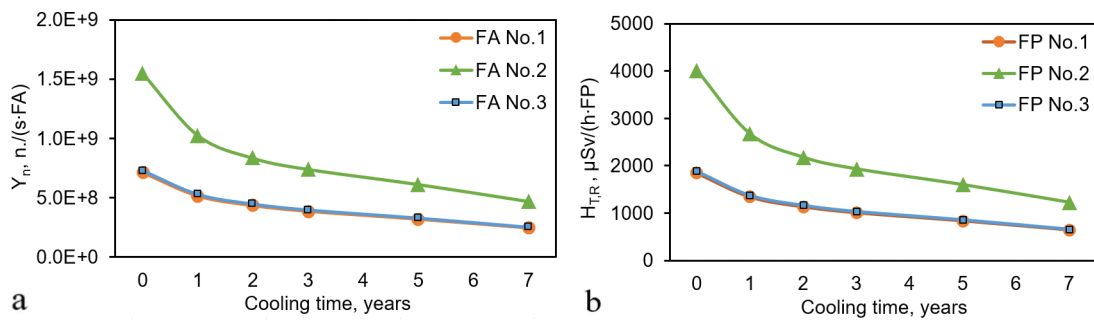


Figure 9. Radiation characteristics depending on the fuel cooling time for three types of FAs: **a.** Intensity of neutron radiation of the FA; **b.** Average value of equivalent dose rate of neutron radiation per one FP.

Table 4. Contribution of the (α , n) reaction to neutron radiation

FA	Units	Beginning of the campaign	End of the campaign	1 year of cooling	2 years of cooling	3 years of cooling	5 years of cooling	7 years of cooling
No. 1	%	9.93	6.05	2.66	1.37	1.39	1.57	1.88
	n./(s·FA)	$5.93 \cdot 10^2$	$4.30 \cdot 10^7$	$1.37 \cdot 10^7$	$5.96 \cdot 10^6$	$5.38 \cdot 10^6$	$5.03 \cdot 10^6$	$4.61 \cdot 10^6$
No. 2	%	99.70	7.39	3.31	1.54	1.53	1.70	1.99
	n./(s·FA)	$2.13 \cdot 10^6$	$1.15 \cdot 10^8$	$3.40 \cdot 10^7$	$1.29 \cdot 10^7$	$1.13 \cdot 10^7$	$1.04 \cdot 10^7$	$9.34 \cdot 10^6$
No. 3	%	9.84	5.97	2.62	1.35	1.38	1.55	1.84
	n./(s·FA)	$5.86 \cdot 10^2$	$4.35 \cdot 10^7$	$1.39 \cdot 10^7$	$6.06 \cdot 10^6$	$5.47 \cdot 10^6$	$5.10 \cdot 10^6$	$4.67 \cdot 10^6$

yield values for FA No. 1 are comparable to the results of the study by Petrovskiy et al. (2020), in which the neutron yield of VVER-1200 spent FA after 3 years of cooling was estimated as $4.66 \cdot 10^8$ n./(s·FA).

Research, including those by Okumura et al. (2000); Shamanin et al. (2010); Pisarev and Kolesov (2020), showed that the main contribution to the calculation uncertainty could come from the uncertainty in determining the nuclear concentrations of the following isotopes: $^{238,239,240,241,242}\text{Pu}$, ^{237}Np , $^{241,242\text{m}}\text{Am}$, $^{242,244,246}\text{Cm}$; many modern program codes including WIMS-D5B, could underestimate the concentration values of minor actinide nuclei (Np, Am, Cm) by 10–20%. Additional uncertainty comes from calculations of neutron yields using SOURCES-4C – judging by the data presented in Figure 1, the values are overestimated by an average of 15%. Therefore, the overall uncertainty of the found values of neutron yields and equivalent dose rates can be approximately $\pm 15\%$. Nevertheless, at the stage of preliminary assessments, the results can be considered quite reliable, since both WIMS-D5B and SOURCES-4C are verified computational programs.

Conclusions

We carried out a computational assessment of the neutronic and radiation characteristics of VVER-1200 FAs of various types. The isotopic composition of α -emitters and neutron emitters has been studied. The main attention was paid to the study of the UO_2 composition with a heterogeneous arrangement of AmO_2 microcapsules. As has been studied by other authors, the thermal conductivity of such fuel does not decrease, as occurs in the case of homogeneous analogues with Gd. Moreover, such fuel is

placed in all fuel rods without changing the enrichment, and therefore does not affect the power peaking in the FA.

The results of neutronic calculations confirm the possibility of using AmO_2 as a BA, since its presence reduces excess reactivity without affecting the duration of the reactor campaign. However, a run-out of reactivity, as in the case of Gd, is not observed due to a more uniform distribution of the BA in the FA and slower burnup due to the smaller macroscopic neutron absorption cross section, therefore the change in $k_{inf}(t, z)$ is close to linear.

Using the SOURCES-4C code, neutron energy spectra were obtained, on the basis of which the neutron components of the radiation characteristics of fresh and spent fuel of three types of FAs were assessed – the intensity of the neutron radiation of the FA and the equivalent dose rate of the neutron radiation of the FP. In fresh fuel containing AmO_2 , the neutron yield is higher compared to Am-free fuel and is formed mainly by (α , n) reactions in AmO_2 microcapsules. In addition, such fuel requires a longer cooling time, since some Am nuclei capture neutrons during reactor operation, which leads to the formation of Cm isotopes through successive nuclear transformations and, accordingly, to a greater neutron yield of SNF. Therefore, to use americium as a possible alternative to gadolinium, it is necessary to develop procedures and regulations for handling the new fuel during its manufacture and after irradiation in the reactor. At the same time, radiation safety must be justified taking into account also beta and gamma emissions from spent fuel isotopes, by assessing their impact on the equivalent dose rate. Although only neutron emissions are included in the equivalent dose rate calculations, this work contributes to the study of the neutronic and radiation characteristics of Am-containing fuel that has potential for use in modern thermal reactors.

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