

MCNP AND ORIGEN CODES VALIDATION BY CALCULATING RBMK SPENT NUCLEAR FUEL ISOTOPIC COMPOSITION*

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Benchmark calculations of RBMK reactor spent nuclear fuel isotopic composition were performed using the MCNP and ORIGEN codes system. The RBMK assembly is explicitly modelled in order to investigate the differences of spent nuclear fuel burn-up due to different coolant density and the fuel rod position (inner or outer ring in the assembly). The modelling results are compared with experimentally measured data of RBMK-1000 fuel isotopic composition with 2% enrichment ²³⁵U fuel at different fuel burn-up. The comparison of the experimentally measured and calculated isotopic composition for the main actinides and fission products allowed validation of MCNP and ORIGEN codes for RBMK spent nuclear fuel calculations. Variations of RBMK spent nuclear fuel burn-up and plutonium isotopic ratios depending on fuel location in the assembly were determined.

Keywords: RBMK reactor, spent nuclear fuel, MCNP5 benchmark calculations, plutonium isotopic ratios

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1. Introduction

Currently used calculation codes and nuclear data for nuclear fuel isotopic composition modelling are well validated for conventional pressurized water (PWR) or boiling water (BWR) nuclear reactors. On the other hand, the computational tools for proper calculation of graphite moderator reactors, one of which is RBMK-1500, are difficult to adapt due to complicated structures, different materials used, and particular physical processes [1, 2]. The validation of the modelling tools is usually done referring to the indirect measurements of spent nuclear fuel (SNF) samples [3, 4]. In addition, there are only a few direct experimental data available for the validation of RBMK type reactor SNF composition and the accessibility of the published results is limited. The detailed report on RBMK-1000 spent nuclear fuel isotopic composition including experimental data and simulations (with WIMS-D4 and CACH2 codes) was done by Kurchatov Institute (Russia) [5], and these data were used in our benchmark calculations using the MonteBurns code system [6].

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2. Modelling tools and procedure

For benchmark calculations of RBMK spent nuclear fuel isotopic composition the MonteBurns code system was used [6] – an automatic-cyclic coupling of the MCNP [7] and ORIGEN2 [8] codes. MonteBurns was successfully benchmarked earlier by simulating the uranium based fuel cycle of the high flux reactor at ILL Grenoble [9] and GT-MHR (Gas Turbine – Modular Helium cooled Reactor) in the case of plutonium (uranium free) fuel cycle [10].

The advantage of the MonteBurns code is that it allows the use of different nuclear data libraries as long as they are compatible with the MCNP format. The ENDF/B-VI data library (as the one most frequently employed with MCNP) was used for the fuel and structure materials, and the JENDL-3.2 data files (having the largest number of fission products) were employed for fission products.

The Monte Carlo N-Particle code MCNP is applied for neutron transport calculation. In brief, particle transport using the Monte Carlo technique is based on the explicit tracking of particles following each particle from a source throughout its life to its death (by parasitic absorption or escape). Probability distributions are randomly sampled using transport data. In

calculations these distributions are used to determine the type of interaction, energy of particles if it scatters, leakage of particles, and the number of neutrons produced if fission occurs. The Monte Carlo method is well suited for solving complicated three-dimensional problems. In our calculations MCNP was used to obtain neutron flux in the fuel assembly. For the input data the MCNP code requires nuclear data, materials, and detailed geometry description.

The neutron spectrum in the fuel assembly changes considerably during the fuel burn-up. Therefore, the evolution calculations of fuel isotopic composition have to be performed with corresponding variable neutron fluxes as it is done with Monteburns [6]. In this work the neutron fluxes and cross sections were recalculated every twenty-five days for further ORIGEN burn-up calculations. In ORIGEN the nuclide composition evolution is calculated for the concentration N_i of each isotope in a material which is affected by neutron flux using the equation [8]

$$\frac{dN_i}{dt} = -\left(\lambda_i + \int \varphi(E, t) \sigma_i(E) dE\right) N_i + \sum_{j \neq i} \left(\lambda_{ji} + \int \varphi(E, t) \sigma_{ji}(E) dE\right) N_j, \quad (1)$$

where λ_i are decay constants of the i th isotope, λ_{ji} are the partial decay constants of the j th isotope to the i th isotope, $\varphi(E, t)$ is the magnitude of the particle flux of energy E , σ_i are the neutron absorption cross sections of the i th isotope, and σ_{ji} are the neutron cross sections for transmutation from isotope j to i .

3. Details of RBMK SNF assembly modelling

The modelling of RBMK reactor fuel isotopic composition was performed according to the experiment [5] done for RBMK-1000 charged with 2% ^{235}U enrichment fuel. In order to obtain the isotopic composition of RBMK-1000 SNF during the experiment [5], several fuel assemblies of different burn-up were cut off at places specified by height and by fuel pellet location (in the inner or outer rings) as it is presented in Table 1. Isotopic composition of U, Pu, Np, Am, Cs, and Nd was obtained after radiochemical separation by applying alpha, gamma, and mass spectrometry methods. The uncertainties of the obtained values were estimated and presented in [5]. The burn-up of the spent nuclear fuel in the samples was evaluated according to the fission products ^{137}Cs , $^{145+146}\text{Nd}$, and ^{148}Nd isotopes concentrations.

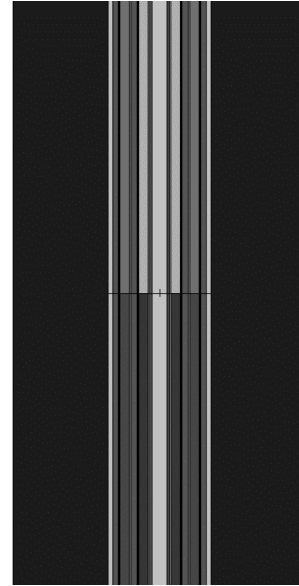


Fig. 1. The fragment of a fuel assembly axially divided according to different coolant densities.

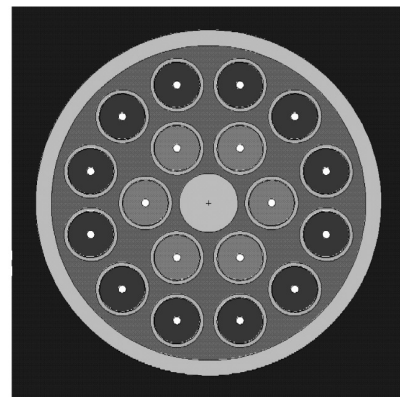


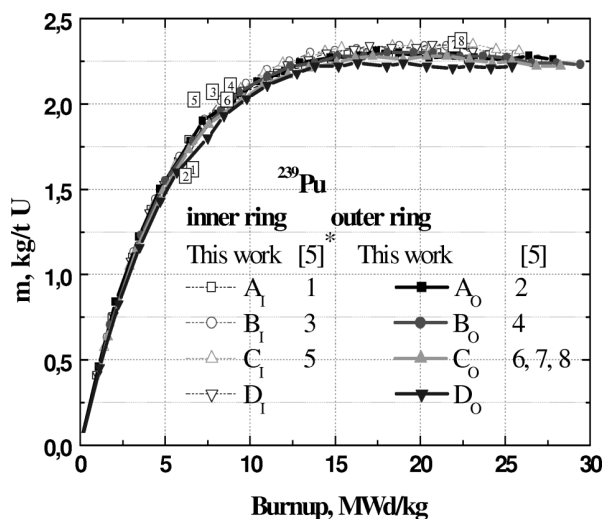
Fig. 2. Cross-sectional view of a fuel assembly with inner and outer fuel pellet rings. The fuel assembly is surrounded by fuel channel wall of Zr (+2.5% Nb) and by graphite moderator.

Table 1. Characteristics of the 2% enriched ^{235}U RBMK-1000 SNF samples [5].

Experimental points	Fuel pellet ring	Burn-up, MW·d/kg	h , cm
1	inner	6.62	585
2	outer	6.2	585
3	inner	7.79	370
4	outer	8.86	370
5	inner	6.7	330
6	outer	8.65	330
7	outer	22	271
8	outer	22.4	256

Table 2. Notation of axially divided regions of fuel assembly for inner and outer fuel pellets rings.

Notation of fuel assembly regions for inner (I) and outer (O) fuel rod rings		h , cm	$\rho(\text{H}_2\text{O})$, g/cm ³
A _I	A _O	686–514.5	0.25
B _I	B _O	514.5–343	0.33
C _I	C _O	343–171.5	0.55
D _I	D _O	171.5–0	0.75

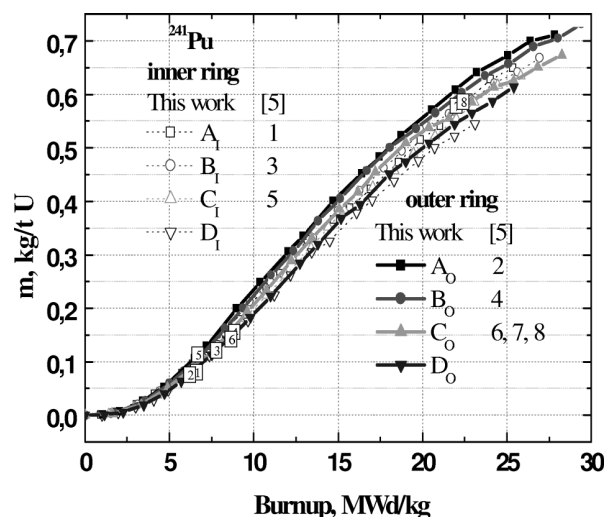
Fig. 3. ²³⁹Pu production in the RBMK reactor fuel assembly as a function of fuel burn-up at different neutron environment conditions, and experimental results [5].

The cross-sectional and longitudinal view of the fuel assembly modelled with MCNP [7] is presented in Figs. 1 and 2. In order to take into account the axial and radial burn-up profile in the RBMK reactor active core the fuel assembly in the MCNP model was divided into 4 regions with different coolant (H₂O) densities (0.25–0.75 g/cm³) as presented in Table 2. The coolant density profile was taken from [5]. The inner and outer fuel pellet rings were separated in the model in order to investigate the influence of different neutron environment (harder or softer neutron spectrum) in different fuel assembly places and in order to compare the modelling results with the experimental values.

4. Results

4.1. Comparison of experimental and modelling results for RBMK SNF

We performed our calculations by simulating one fuel assembly using periodical boundary conditions at the constant 3 MW power (the maximal 5000 MW RBMK reactor power). The first intention was to vali-

Fig. 4. Evolution of ²⁴¹Pu mass in the RBMK reactor fuel assembly at different assembly height for two fuel pellets rings, and experimental results [5].

date the MCNP+ORIGEN codes system for the RBMK reactor calculations. It was done by comparing experimental RBMK-1000 fuel assembly isotopic composition measurements with modelling results of 2% enrichment fuel isotopic composition. The fuel evolution in the RBMK fuel assembly during 1000 days was simulated and burn-up dependent mass evolution curves for actinides (Pu, Am, Cm) and some fission products (Cs and Nd) were plotted. Comparison of the modelling curves against experimental results for ²³⁹Pu, ²⁴¹Pu, ²⁴⁴Cm, and ¹⁴⁸Nd is presented in Figs. 3–6. The mass evolution of ²³⁹Pu in a different neutron environment (about 5% harder spectrum in the inner fuel pellet ring and in the upper part of the assembly) is very similar in all the burn-up range considered, as shown in Fig. 3. The experimental data from different burn-up samples are in the acceptable ranges compared with modelling results for ²³⁹Pu. Modelling results were found to be in good agreement with experimental data also for the other actinides and fission products (see Figs. 4–6). The production of ²⁴¹Pu in the SNF can vary up to 5% for different fuel assembly positions at different fuel burn-up. Variations in thermal neutrons spectrum affect the formation of ²⁴⁰Pu and subsequently the formation of ²⁴¹Pu by neutron capture reaction. The largest (a few times) differences between the experimental data and calculations were obtained for ²⁴⁴Cm, which, due to its formation in different chain reactions, is extremely sensitive even to small variations of neutron flux and spectrum. In addition, this difference also might appear due to inadequacy of data libraries used in the case of minor actinides. The excellent agreement between the ¹⁴⁸Nd exper-

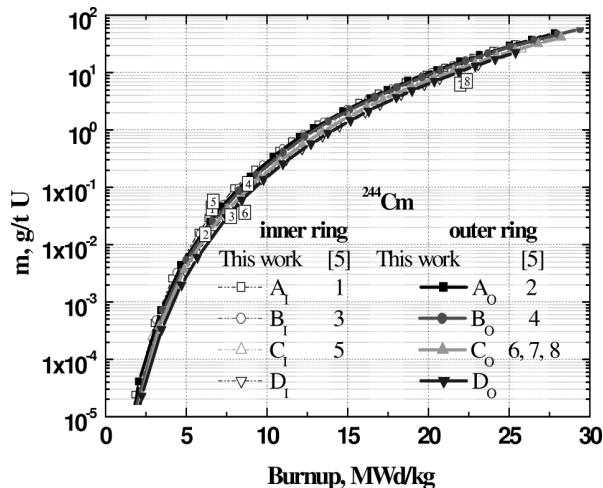


Fig. 5. ^{244}Cm mass as a function of burn-up in the RBMK fuel assembly for four coolant densities and two fuel pellets rings, and the experimental results [5].

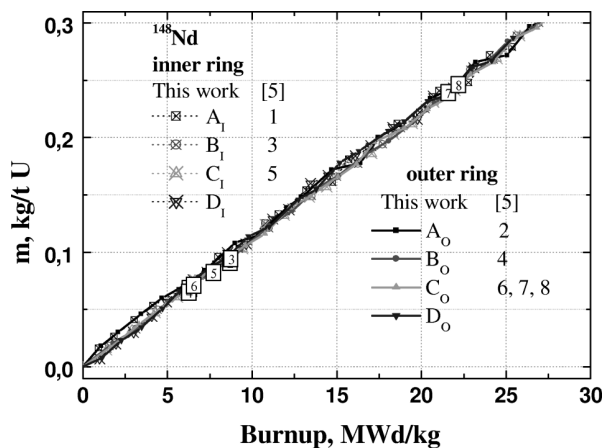


Fig. 6. ^{148}Nd mass as a function of burn-up in the RBMK fuel assembly for four coolant densities and two fuel pellets rings, and the experimental results [5].

imental data and modelling of ^{148}Nd mass evolution as a function of burn-up has confirmed the reliability of the MCNP+ORIGEN codes system for calculation of fission products of RBMK SNF. These results allow us to conclude that numerical calculations of RBMK SNF made using the MCNP+ORIGEN codes system are in a good agreement with experimental data [5] for the main actinides characterizing nuclear waste ^{239}Pu , ^{241}Pu , ^{244}Cm , and the fission products.

4.2. Spent nuclear fuel burn-up variations depending on fuel location

The detailed modelling of the RBMK reactor fuel assembly, including coolant density profile (4 regions with densities of 0.25–0.75 g/cm³) and two separate fuel pellet rings, allowed us to investigate the axial and

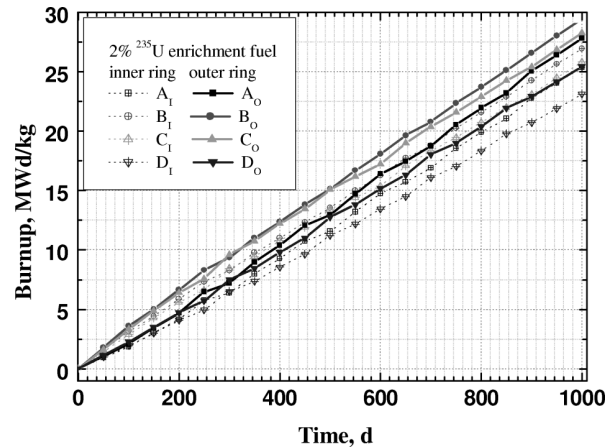


Fig. 7. The RBMK reactor fuel burn-up dependence on the irradiation time and the fuel burn-up variations due to particular fuel pellet location.

radial burn-up variations at the different places of the fuel assembly. Eight different fuel burn-up curves in Fig. 7 correspond to four fuel assembly regions characterized by variable coolant density (see Table 2 for notations) and to two rings with inner and outer fuel pellets. It is clearly seen that the fuel burn-up in the inner ring of the fuel pellets is lower compared to the outer fuel ring. The differences in burn-up are about 10% for each coolant density region after 1000 days of irradiation. The maximal differences of 22% in fuel burn-up are observed between the bottom (D₁ at 171.5–0 cm height, with $\rho(\text{H}_2\text{O}) = 0.75 \text{ g/cm}^3$) inner ring fuel and the middle-upper (B₀ at 514.5–343 cm height, with $\rho(\text{H}_2\text{O}) = 0.33 \text{ g/cm}^3$) outer ring fuel. The reason for such differences is the neutron flux variations and the heterogeneous neutron absorption, because harder neutron spectrum is in the inner fuel pellet ring of the assembly and more neutrons are absorbed in the lower part of the assembly due to higher density of the water (less neutrons are available for fission reaction).

4.3. Plutonium isotopic ratios for 2% enrichment RBMK fuel in different neutron environment

The different fuel position in the fuel assembly leads to different plutonium isotopic ratios, which serve for radioactive nuclear waste and contamination identification (military or civil plutonium, reactor type RBMK or PWR, etc.). Applying alpha spectrometry analysis one is able to obtain the ratio of $^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$. This characteristic isotopic ratio is presented in Fig. 8 as a function of irradiation time in the same fuel assembly but different fuel pellet position in the assembly. One can notice that this parameter strongly depends on the irradiation time. The $^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$ isotopic

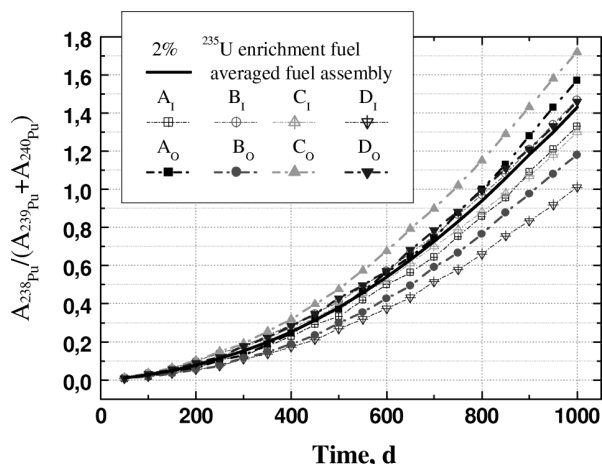


Fig. 8. $^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$ activity ratio as a function of time in the same fuel assembly but for different fuel pellet position in the assembly.

ratio calculated for the averaged fuel assembly fits the ratios calculated in the middle region of the fuel assembly (B_I and C_O), but large 18–32% deviations are observed for the bottom part of the assembly D_I, D_O with 0.75 g/cm³ water density and also about 22% in the upper part B_O with 0.33 g/cm³ water density. Depending on the irradiation time and on the fuel pellet locations in the assembly the $^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$ isotopic ratio can vary up to 30%. Similar conclusions can be drawn concerning the other Pu isotopic characteristic ratio $^{240}\text{Pu}/^{239}\text{Pu}$ that can be measured by gamma or mass spectrometry methods. The variation of the $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio due to different fuel pellet position in the assembly at different fuel irradiation time is shown in Fig. 9. It can be seen that the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio is less dependent on fuel position but still the deviation from the averaged fuel assembly curve makes up to 12% (maximal deviation in the D_I case).

On the contrary, the relation of isotopic mass ratios $^{242}\text{Pu}/^{239}\text{Pu}$ and $^{240}\text{Pu}/^{239}\text{Pu}$ calculated for different burn-up does not depend on different fuel position in the assembly as shown in Fig. 10. The maximal deviation of the ratios in different fuel places is less than 5%. This functional relation can be used for identification of RBMK reactor SNF.

5. Conclusions

The benchmark calculations of RBMK reactor spent nuclear fuel isotopic composition using MCNP and ORIGEN codes were performed. The modelling results were compared with experimentally measured data of the RBMK-1000 fuel isotopic composition with 2% en-

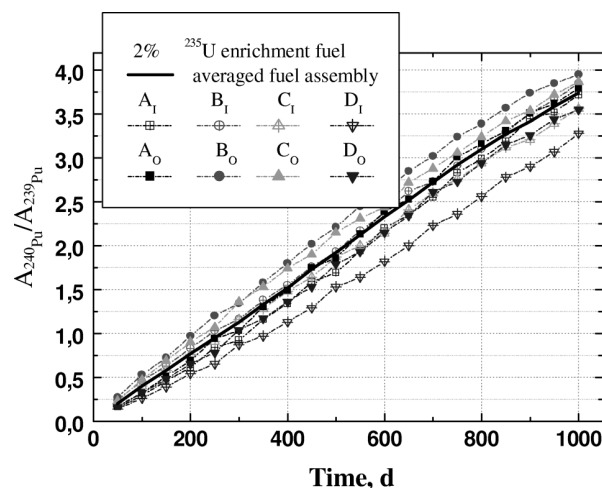


Fig. 9. Variation of $^{240}\text{Pu}/^{239}\text{Pu}$ activity ratio due to different fuel pellet position in the assembly at different fuel irradiation time.

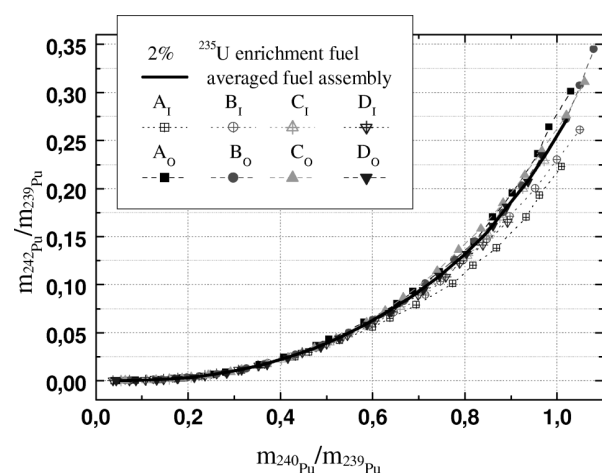


Fig. 10. Comparison of $^{242}\text{Pu}/^{239}\text{Pu}$ and $^{240}\text{Pu}/^{239}\text{Pu}$ mass ratios for RBMK reactor SNF.

richment ^{235}U fuel at different fuel burn-up. Modelling results were found to be in good agreement with experimental data for the main actinides characterizing nuclear waste: ^{239}Pu , ^{241}Pu , ^{244}Cm . The largest differences between experimental data and calculations were observed for ^{244}Cm because its formation via different reactions is extremely sensitive even to small neutron flux and spectrum variations. The excellent agreement between ^{148}Nd experimental data and modelling of ^{148}Nd mass evolution as a function of burn-up has confirmed the reliability of the MCNP+ORIGEN codes system for the calculation of fission products of RBMK reactor SNF.

The RBMK reactor assembly was explicitly modelled including inner and outer fuel pellet rings and with vertically varying coolant density along the fuel channel in order to investigate the influence of fuel location (and consequently different neutron environ-

ment) on spent nuclear fuel burn-up and plutonium isotopic ratios. The differences in burn-up are about 10% for each coolant density region. The maximal difference of 22% in fuel burn-up is observed between the bottom part of the assembly inner ring fuel and the middle-upper part of the assembly outer ring fuel. It is due to the harder neutron spectrum in the inner fuel pellet ring and respectively fewer neutrons available in the lower part of the assembly. It has been found that the production of ^{241}Pu in the SNF, depending on the fuel burn-up, can vary up to 5% for different fuel assembly positions.

Analysis of the plutonium isotopic ratios has shown that:

- depending on irradiation time and on fuel pellet location in the assembly the alpha activity ratio of Pu isotopes $^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$ can vary up to 30%;
- the isotopic $^{240}\text{Pu}/^{239}\text{Pu}$ ratio is less dependent on fuel pellet position but still the deviation from the averaged fuel assembly makes up to 12%;
- the ratio $^{242}\text{Pu}/^{239}\text{Pu}$ against $^{240}\text{Pu}/^{239}\text{Pu}$ is stable within 10% for different burn-up independently of different fuel position.

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MCNP IR ORIGEN PROGRAMŲ PATIKRA RBMK REAKTORIAUS PANAUDOTO BRANDUOLINIO KURO IZOTOPINĖS SUDĖTIES VERTINIMUI

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Santrauka

Dabartinės kompiuterinės panaudoto branduolinio kuro sudėties reaktoriuje modeliavimo programos bei jose naudojami įvertintieji branduoliniai duomenys yra gerai pritaikyti labiausiai paplitusiems pasaulyje suslėgto (PWR) arba verdančio (BWR, VVER) lengvojo vandens reaktorių parametrams skaičiuoti. Nevienalyčiams reaktoriams su grafito lėtikiu, tokiems kaip RBMK-1500, dėl skirtingos geometrinės sandaros ir medžiagų sudėties bei fizikinių vyksmų savitumo, egzistuojančios programos yra sunkiai arba tik dalinai pritaikomos [1, 2]. Patikrintas MCNP ir ORIGEN programų tinkamumas RBMK reaktoriaus panaudoto branduolinio kuro izotopinės sudėties skaičiavimams. Modeliavimo rezultatai palyginti su eksperimentiniais RBMK-1000 kuro izotopinės sudėties duomenimis 2% ²³⁵U išodrinimo kurui [5]. Parodyta, kad pagrindiniams branduolines atliekas apibūdinantiems ²³⁹Pu, ²⁴¹Pu, ²⁴⁴Cm nuklidams modeliavimo duomenys gerai sutampa su eksperimentiškai išmatuotais. Didžiausias skirtumas pastebėtas ²⁴⁴Cm, kadangi šio nuklido susidarymas labai priklauso nuo daugelio tarpinių produktų, jautrių neutronų spektro pasikeitimams. Modeliavimo ir eksperimentiniai rezultatai labai gerai sutapo dalijimosi produktams (Nd).

Remiantis gautais rezultatais, galima teigti, kad MCNP+ORIGEN programų sistema tinka RBMK panaudotam branduoliniam kurui (PBK) apibūdinti. Taip pat ištirta PBK išdegimo bei plutonio izotopinių santykių priklausomybė nuo kuro tablečių padėties rinklėje, kai modeliuojant atsižvelgiama į skirtingą vandens tankį rinklėje ir kuro tablečių išsidėstymą atitinkamai vidiniame arba išoriniame kuro elementų žiede. RBMK kuro rinklės modeliavimo su išoriniu ir vidiniu kuro elementų žiedais bei keturiais skirtingais vandens tankio regionais rezultatai parodė, kad kuro išdegimas gali skirtis iki 10% kiekvienam skirtingo vandens tankio regionui rinklėje, o didžiausias 22% išdegimo skirtumas rastas tarp apatinės rinklės dalies vidinio ir vidurinės–viršutinės dalies išorinio kuro elementų žiedo. Plutonio izotopinių santykių analizė parodė, kad, priklausomai nuo kuro tablečių padėties kuro rinklėje, ²³⁸Pu/(²³⁹Pu+²⁴⁰Pu) aktyvumų santykis gali kisti iki 30%. ²⁴⁰Pu/²³⁹Pu aktyvumų santykis yra mažiau jautrus kuro tablečių padėčiai: nukrypimas nuo vidutinės kuro rinklės yra iki 12%. ²⁴²Pu/²³⁹Pu ir ²⁴⁰Pu/²³⁹Pu masių santykių kreivė, pagal kurią galima atskirti RBMK kurą nuo kitų tipų reaktorių kuro, praktiškai nepriklauso nei nuo kuro išdegimo, nei nuo kuro tabletės padėties rinklėje.