

# A study into the propagation of the uncertainties in nuclear data to the nuclear concentrations of nuclides in burn-up calculations\*

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## Abstract

The key papers on estimating the uncertainties in nuclear data deal with the influence of these uncertainties on the effective multiplication factor by introducing the so-called sensitivity factors and only some of these are concerned with the influence of such uncertainties on the life calculation results. On the other hand, the uncertainties in reaction rates, the neutron flux, and other quantities may lead to major distortions in findings, this making it important to be able to determine the influence of uncertainties on the nuclear concentrations of nuclides in their burn-up process. The possibility for the neutron flux and reaction rate uncertainties to propagate to the nuclear concentrations of nuclides obtained as part of burn-up calculations are considered using an example of a MOX-fuel PWR reactor cell. To this end, three burn-up calculation cycles were performed, and the propagation of uncertainties was analyzed. The advantages of the uncertainty estimation method implemented in the VisualBurnOut code consists in that all root-mean-square deviations are obtained as part of one calculation as the statistical method, e.g. GRS (Generation Random Sampled), requires multiple calculations. The VisualBurnOut calculation results for the root-mean-square deviations in nuclear concentrations were verified using a simple model problem. It is shown that there is a complex dependence of the propagation of the root-mean-square deviations in the nuclear concentrations of nuclides in the process of fuel burn-up, and, therefore, further studies need to aim at investigating the influence of uncertainties in nuclear data on the nuclear concentrations of nuclides.

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## Keywords

Reactor plant, burn-up calculations, uncertainties in nuclear data, uncertainties in nuclear concentrations of nuclides, Monte Carlo method

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## Introduction

At the present time, a great deal of attention is given to estimating the influence of uncertainties in nuclear data

on the parameters of different reactor plants. Largely, however, the influence of uncertainties in nuclear data on the key parameters ( $k_{\text{eff}}$ , reaction rates, and others), are studied using sensitivity factors. Thus, estimated un-

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certainties for different thermal reactors with a burn-up of 40 to 100 MW·day/kg equal 0.5 to 2% for  $k_{\text{eff}}$  (Aliberti et al. 2006, OECD/NEA 2008, Rochman et al. 2012, Wieselquist et al. 2013), 1 to 5% for Doppler reactivity coefficient (Aliberti et al. 2006, OECD/NEA 2008), and 1 to 10% for decay heat (Aliberti et al. 2006, OECD/NEA 2008, Rochman et al. 2012, Wieselquist et al. 2013, Stover 2007).

Nuclear data for high energies are less accurate, and estimated uncertainties for fast reactors have, accordingly, higher values than those for thermal reactors, e.g., 1 to 4% for  $k_{\text{eff}}$  (Aliberti et al. 2006, OECD/NEA 2008, Stover 2007, Rochman et al. 2011), 3 to 8% for temperature reactivity coefficient (Aliberti et al. 2006, OECD/NEA 2008), 10 to 20% for sodium void reactivity coefficient (OECD/NEA 2008, Rochman et al. 2011), and 1 to 20% for decay heat. To the maximum extent, poor accuracy of data affects accelerator-driven systems used for actinide burning, while uncertainties for different quantities may reach tens of percent (OECD/NEA 2008, Aliberti et al. 2004).

Only several works deal with the effects of uncertainties in nuclear data on the nuclear concentrations of nuclides obtained in the process of calculating the fuel burn-up in the reactor (Gandini et al. 1977, Usachev et al. 1979, Kolesov et al. 2017, Garcia-Herranz et al. 2008, Takeda et al. 1999, Tohjoh et al. 2006, Park et al. 2011, Quentin Newell and Charlotta Sanders 2015, Rochman et al. 2014, Rochman et al. 2012, Andrew Conant et al. 2017, Da Cruz et al. 2014, Sjostrand et al. 2013). The first attempts to estimate the effects of uncertainties in nuclear data on the concentrations of nuclides in the process of burn-up appear to have been made in (Gandini et al. 1977, Usachev et al. 1979).

Quantitatively estimating the uncertainties in concentrations as part of burn-up calculations will allow a better prediction of the fuel's isotopic composition for optimization tasks of transmutation, recycling, and waste removal. Transmutation indices depend greatly on nuclear data, and it is important to quantitatively estimate the resulting uncertainties in integral parameters of fast reactor systems, since their fuel may be heavily saturated with minor actinides, such as americium and curium, the cross-sections (reaction rates) of which are poorly known. Knowing the isotopic composition during the reactor operation is essential for estimating the long-term radiotoxicity and decay heat of spent fuel, changes in the reactivity margin, and the growth in the pressure of gas and in the concentration of volatile fission products (which form the source term in emergencies) (Sjostrand et al. 2013). Important for the spent fuel transportation and storage are both accurate calculation of the total decay heat and the uncertainty in  $k_{\text{eff}}$ . Solving fuel burn-up problems is also important for estimating the accumulation of certain isotopes generated in reactor plants, such as plutonium isotopes. And it is rather hard in most cases to determine experimentally their concentrations in spent fuel. Currently, therefore, of a major importance are numerical methods for solving burn-up problems. On the other hand, uncertainties in estimated reaction rates, neutron fluxes, and other quantities

can lead to major distortions in findings, which makes it important to know how to determine the effects of such uncertainties on the nuclear concentrations of nuclides in the process of fuel burn-up.

## Problem definition

Let  $\mathbf{N}(t) = [N_1(t), N_2(t), N_M(t)]^T$  be the vector of nuclear concentrations consisting of  $M$  different nuclides at the time  $t$ . The time-dependent change in the nuclear concentrations in the neutron field is described by a system of differential equations

$$d\mathbf{N}/dt = \mathbf{A}\mathbf{N} = \boldsymbol{\lambda}\mathbf{N} + \boldsymbol{\sigma}^{\text{eff}}\Phi\mathbf{N}, \quad (1)$$

where  $\mathbf{A}$  is the matrix of the transitions of certain nuclides to others due to nuclear reactions and decays;  $\boldsymbol{\lambda}$  is the matrix of the values for the decay constants of the size  $M \times M$ ;  $\boldsymbol{\sigma}^{\text{eff}}$  is the matrix of single-group neutron cross-sections; and  $\Phi$  is the averaged integral neutron flux. Taking into account that  $\mathbf{N}_0 = \mathbf{N}(0)$  is the initial vector of nuclear concentrations, we shall write down the solution as

$$\mathbf{N}(t) = \exp(\mathbf{A}t)\mathbf{N}_0 \quad (2)$$

with the constant neutron spectrum (therefore, with constant single-group microscopic cross-sections and the constant neutron flux) throughout the time step  $[0, t]$ .

Nuclear systems, in which changes in the isotopic composition of fuel affect the neutron flux distribution, require a series of combined neutronic and fuel burn-up calculations. Such combined calculations have the entire burn-up cycle divided into several successive time intervals. The ionizing radiation transport is calculated for each time interval, and the reaction rates and the neutron flux are also calculated which are used to solve the burn-up equation and to obtain the isotopic composition of fuel at the respective time interval end.

The purpose of the work is not only to calculate the vector  $\mathbf{N}$  of the time-dependent nuclear isotope concentration but also to estimate the effects of different uncertainty sources arising as the result of a combined burn-up calculation of nuclear concentrations.

## Sources of uncertainties in burn-up calculations

Let there be no uncertainty in the initial nuclear concentrations of nuclides though it is possible to take these into account as well. Uncertainties can be found in all the parameters a burn-up equation includes, i.e., in decay constants  $\boldsymbol{\lambda}$ , in single-group effective microscopic cross-sections  $\boldsymbol{\sigma}^{\text{eff}}$ , and in the integral neutron flux  $\Phi$ :  $\mathbf{N} = \mathbf{N}(\boldsymbol{\lambda}, \boldsymbol{\sigma}^{\text{eff}}, \Phi, t]$  whence it follows that  $\Delta\mathbf{N}$  depends on  $\Delta\boldsymbol{\lambda}$ ,  $\Delta\boldsymbol{\sigma}^{\text{eff}}$ ,  $\Delta\Phi$ , where the symbol  $\Delta$  means the uncertainty or the relative error of the respective value.

1. Uncertainties in decay constants  $\Delta\lambda$  are normally given in evaluated nuclear data libraries.
2. Uncertainties in single-group effective microscopic cross-sections  $\Delta\sigma^{eff}$  depend both on uncertainties in evaluated nuclear data of microscopic cross-sections  $\Delta\sigma^g$  and on uncertainties in the neutron flux  $\Delta\Phi^g(E)$  (the index  $g$  here means the energy group number). Knowing the covariance matrices which can be obtained from respective files of evaluated neutron data, one can obtain uncertainties in single-group effective microscopic cross-sections  $\Delta\sigma^{eff}$ , e.g. using the ERRORJ module of the NJOY software package.

An analysis of uncertainties in the nuclear data available in the latest international nuclear data libraries has shown that:

- there is no covariance data of important nuclides;
- the question of the covariance data reliability remains open.

In this case, the results obtained using this data should be viewed as a sort of a “proof of principle”; calculations should be repeated up to obtaining more accurate data. On the other hand, uncertainties in the neutron flux are explained by uncertainties in microscopic cross-sections and nuclear concentrations of nuclides (generally, in all input data required for the transport equation) and by the statistical nature of the neutron transport Monte Carlo calculation as such.

3. Uncertainties in the integral neutron flux  $\Delta\Phi$ . A normalizing coefficient is required to obtain the neutron flux value. As a rule, constant power is assumed to be such a factor, that is, there is a control mechanism which will change (compensate for) the neutron flux value to keep the required constant power level. If we designate full power as  $P$ , then

$$P = KN\sigma_f\Phi V, \quad (3)$$

where  $V$  is the core volume;  $N\sigma_f\Phi$  is the fission rate; and  $K$  is the conversion factor. It can be seen from the equation that the uncertainty in the integral neutron flux will depend on the uncertainties in the nuclear concentrations of nuclides and uncertainties in the single-group microscopic cross-sections of the fissile material.

Therefore, the sources of uncertainties in burn-up calculations can be classified as follows:

- 1) uncertainties in initial nuclear data;
- 2) uncertainties caused by the statistical nature of the neutron transport Monte Carlo calculation;
- 3) uncertainties caused by the normalizing factor.

$$N = N(\lambda, \sigma^{eff}, \Phi, t) = N(\lambda, \sigma^{eff}, \Delta\Phi, t). \quad (4)$$

The paper investigates the effects of uncertainties in the neutron flux and in the reaction rates for different nuclides on burn-up calculations.

The most intuitively understood method to propagate the uncertainties in nuclear data to nuclear concentrations in the process of burn-up consists in selecting, for each calculation, the reaction rates and/or the neutron flux in a statistical (e.g., lognormal) manner with the preset root-mean-square deviation. The calculation is performed repeatedly. The method is simple and versatile but is extremely costly since it often requires a calculation to be repeated hundreds of times. Several patterns were developed to estimate uncertainties with only one set of solutions (Garcia-Herranz et al. 2008, Takeda et al. 1999, Tohjoh et al. 2006, Park et al. 2011) but these were not adapted to a significant extent. We shall thus obtain the respective distributions of nuclear concentrations and shall be able to estimate their root-mean-square deviations.

It was shown in (Kolesov et al. 2017) that such root-mean-square deviations can be estimated with a good accuracy using variations of respective nuclear data (e.g., reaction rates and/or the neutron flux) by any value having the respective response in the nuclear concentrations. Where variations coincide with the respective root-mean-square deviations in the direct statistical approach, then the variations in the nuclear concentrations will coincide with the root-mean-square deviations of the nuclear concentrations. This nuclear concentration uncertainty estimation technique was implemented in the VisualBurnOut code (Kolesov et al. 2009) where no multiple calculations using a statistical approach are required. All nuclear concentration variations are obtained for one VisualBurnOut run. And where constants are recalculated as part of the current step, then all variations of the nuclear concentrations obtained in the previous step need to be included in the calculation of variations for the next step.

## Initial data for calculations

The computational model is a square-shaped PWR cell containing MOX fuel. The geometry and the composition of the fresh fuel, the cladding and the moderator are shown in Tables 1 through 5 and 7. Table 6 presents the temperatures of the materials.

**Table 1.** Geometry of a square three-zone cell.

Pitch, cm	1.3127
External radius, cm	0.475
Cladding thickness, cm	0.065
Fuel pellet radius, cm	0.410

**Table 2.** Isotopic composition of plutonium in fresh MOX fuel.

Isotope	Isotopic composition, % of $Pu_{total}$
$^{238}Pu$	0.05
$^{239}Pu$	93.6
$^{240}Pu$	6.0
$^{241}Pu$	0.3
$^{242}Pu$	0.05

**Table 3.** Isotopic composition of uranium in fresh MOX fuel.

Isotope	Isotopic composition, % of U <sub>total</sub>
<sup>234</sup> U	0.00119
<sup>235</sup> U	0.25000
<sup>238</sup> U	99.74881

**Table 4.** Initial enrichment of MOX fuel.

Content of plutonium in MOX fuel, % of Pu <sub>total</sub> /[U+Pu]	MOX fuel enrichment, % of Pu <sub>fissile</sub> /[U+Pu]
4.377	4.110

**Table 5.** Composition of non-fissile materials.

Isotope	Nuclear concentrations, 10 <sup>24</sup> nuclei/cm <sup>3</sup>
Zircaloy-2 (5.8736 g/cm <sup>3</sup> – reduced density)	
Zr (natural)	3.8657E-2
Fe (natural)	1.3345E-4
Cr (natural)	6.8254E-5
Coolant/moderator (600 ppm of boron; 0.7245 g/cm <sup>3</sup> )	
H	4.8414E-2
O	2.4213E-2
<sup>10</sup> B	4.7896E-6
<sup>11</sup> B	1.9424E-5

**Table 6.** Temperatures of materials.

Material	Temperature, K
Fuel	900
Cladding	620
Coolant/moderator	575

Single-group reaction rates and fluxes calculated by Monte Carlo method were used to calculate the isotope kinetics using the VisualBurnOut code. The calculations considered 254 nuclides for which single-group cross-sections, obtained based on the JENDL-3.2 and JEF-2.2 libraries, were calculated. Each calculation had 1000 neutron generations traced with 1000 neutron histories per generation.

## Calculation results

The following model problem is set to verify the calculations of uncertainties in nuclear concentrations obtained using the VisualBurnOut code: only the fission and radiation capture of two nuclides, <sup>240</sup>Pu and <sup>241</sup>Pu, are considered. Then the system of differential equations is written as

$$\begin{cases} \frac{dN_{40}(t)}{dt} = -\sigma_c^{40}\Phi N_{40}(t) - \sigma_f^{40}\Phi N_{40}(t); \\ \frac{dN_{41}(t)}{dt} = -\sigma_c^{41}\Phi N_{41}(t) - \sigma_f^{41}\Phi N_{41}(t) + \sigma_c^{40}\Phi N_{40}(t). \end{cases} \quad (5)$$

The analytical solution is written as

$$\begin{cases} N_{40}(t) = N_{40}^0 \exp(-(\sigma_c^{40} + \sigma_f^{40})\Phi t); \\ N_{41}(t) = N_{41}^0 \exp(-(\sigma_c^{41} + \sigma_f^{41})\Phi t) + \frac{\sigma_c^{40} N_{40}^0}{\sigma_c^{41} + \sigma_f^{41} - \sigma_c^{40} - \sigma_f^{40}} \times \\ \times [\exp(-(\sigma_c^{40} + \sigma_f^{40})\Phi t) - \exp(-(\sigma_c^{41} + \sigma_f^{41})\Phi t)]. \end{cases} \quad (6)$$

**Table 7.** Initial composition of MOX fuel.

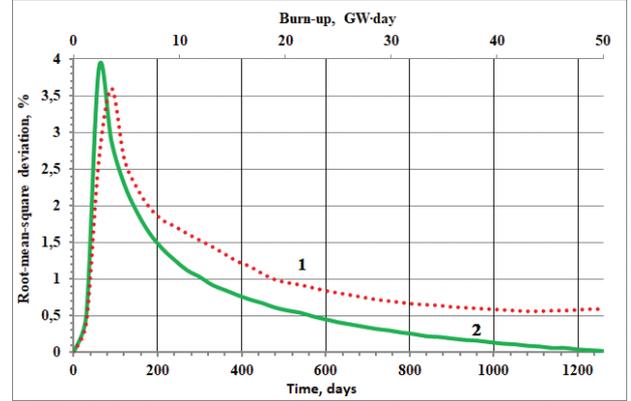
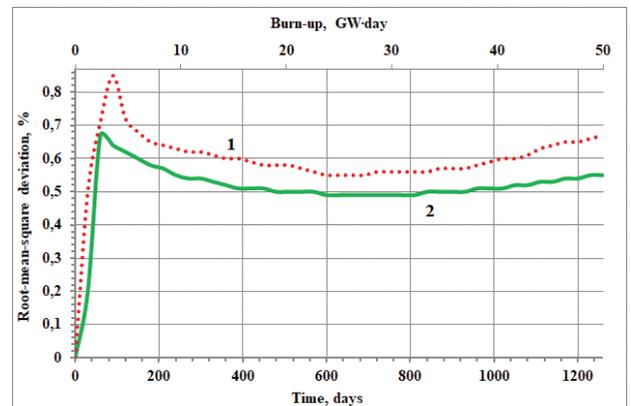
Isotope	Nuclear concentrations, 10 <sup>24</sup> nuclei/cm <sup>3</sup>
<sup>234</sup> U	2.7043E-7
<sup>235</sup> U	5.6570E-5
<sup>238</sup> U	2.2286E-2
<sup>238</sup> Pu	4.5941E-7
<sup>239</sup> Pu	8.5640E-4
<sup>240</sup> Pu	5.4669E-5
<sup>241</sup> Pu	2.7221E-6
<sup>242</sup> Pu	4.5180E-7
<sup>16</sup> O	4.6515E-2

The sensitivity factors with respect to the neutron flux are as follows:

$$\begin{cases} \frac{\partial N_{40}(t)}{\partial \Phi} = -N_{40}^0 (\sigma_c^{40} + \sigma_f^{40}) t \cdot \exp(-(\sigma_c^{40} + \sigma_f^{40})\Phi t); \\ \frac{\partial N_{41}(t)}{\partial \Phi} = -N_{41}^0 (\sigma_c^{41} + \sigma_f^{41}) t \cdot \exp(-(\sigma_c^{41} + \sigma_f^{41})\Phi t) + \frac{\sigma_c^{40} N_{40}^0 t}{\sigma_c^{41} + \sigma_f^{41} - \sigma_c^{40} - \sigma_f^{40}} \times \\ \times [- (\sigma_c^{40} + \sigma_f^{40}) \exp(-(\sigma_c^{40} + \sigma_f^{40})\Phi t) + (\sigma_c^{41} + \sigma_f^{41}) \cdot \exp(-(\sigma_c^{41} + \sigma_f^{41})\Phi t)]. \end{cases} \quad (7)$$

Uncertainties in the nuclear concentrations are further found for the nuclides <sup>240</sup>Pu and <sup>241</sup>Pu:  $\partial N_{40}(t) \Delta \Phi \times 100\% / \partial \Phi N_{40}(t)$  и  $\partial N_{41}(t) \Delta \Phi \times 100\% / \partial \Phi N_{41}(t)$ .

Figs 1, 2 present the root-mean-square deviations in the nuclear concentrations for the nuclides <sup>241</sup>Pu and <sup>240</sup>Pu respectively, as obtained numerically and analytically with a 10% flux perturbation.

**Figure 1.** Root-mean-square deviation in the nuclear concentration of <sup>241</sup>Pu: 1 – analytical calculation; 2 – VisualBurnOut calculation.**Figure 2.** Root-mean-square deviation in the nuclear concentration of <sup>240</sup>Pu: 1 – analytical calculation; 2 – VisualBurnOut calculation.

It can be seen from the figures that the root-mean-square deviations behave in a complicated manner depending on the burn-up time; secondly, the analytically obtained results agree well with the VisualBurnOut numerical calculation results.

Three burn-up cycles were conducted further using the VisualBurnOut code: up to 16, 32 and 48 GW×day/t for a PWR with MOX fuel as described above. The neutron flux for keeping the linear power was recalculated with a step of 30 days which approximately corresponds to 1.1 GW×day/t. Single-group constants were recalculated as part of each burn-up step. We calculated the burn-up defining the root-mean-square deviations for the neutron flux and the reaction rates, and further analyzed the behavior of the nuclear concentration uncertainties depending on time while taking into account both the individual contribution of the uncertainty for each quantity and the joint contribution. The root-mean-square deviations of the burn-up calculation input data amounted to 10%.

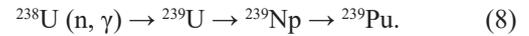
The results of calculating the root-mean-square deviations in the nuclear concentrations of nuclides, depending on uncertainties in various nuclear data, are presented be-

low. It is assumed for all cases that the root-mean-square deviations of the reaction rates and the neutron flux amount to 10%.

Fig. 3 presents the nuclear concentration of  $^{239}\text{Pu}$  as a function of time, and Fig. 4 presents its root-mean-square deviations caused by the uncertainties in the neutron flux and the reaction rates for  $^{238}\text{U}(n,\gamma)$ ,  $^{239}\text{Pu}(n,f)$ , and  $^{239}\text{Pu}(n,\gamma)$ . As shown in Fig. 4, the root-mean-square deviations for the nuclear concentration of  $^{239}\text{Pu}$  are different depending on which of the parameters is perturbed.

Figs 5, 6 present similar data for  $^{241}\text{Pu}$  caused by uncertainties in the neutron flux and the reaction rates for  $^{241}\text{Pu}(n,f)$ ,  $^{241}\text{Pu}(n,\gamma)$ , and  $^{240}\text{Pu}(n,\gamma)$ .

The uncertainty in the  $^{239}\text{Pu}$  nuclear concentration is formed largely by uncertainties in the  $^{239}\text{Pu}$  fission reaction and  $^{238}\text{U}$  radiation capture rates. This can be explained by the fact that  $^{239}\text{Pu}$  is obtained from  $^{238}\text{U}$  in the chain of the following transformations:



The major contributors to the uncertainty in the nuclear concentration of  $^{241}\text{Pu}$  are uncertainties in the  $^{241}\text{Pu}$  fission

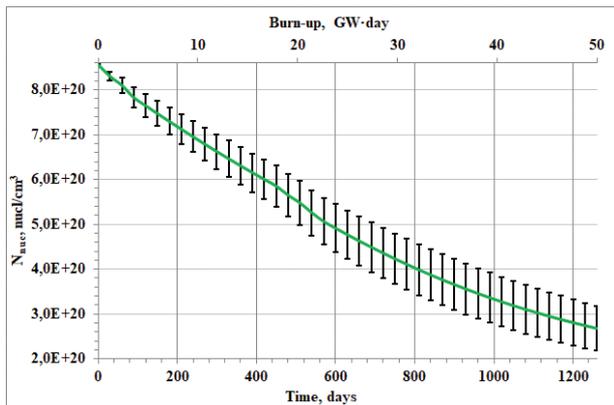


Figure 3. Nuclear concentration of  $^{239}\text{Pu}$ .

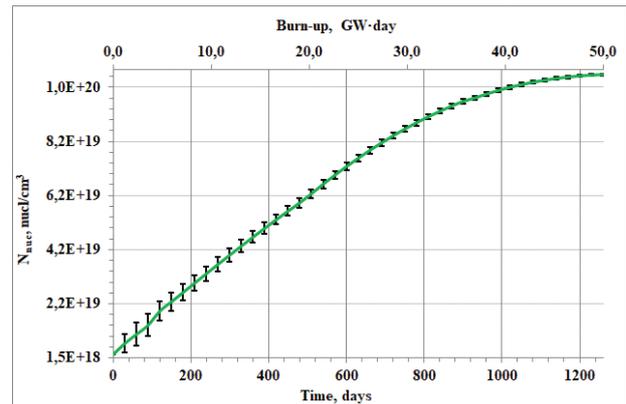


Figure 5. Nuclear concentration of  $^{241}\text{Pu}$ .

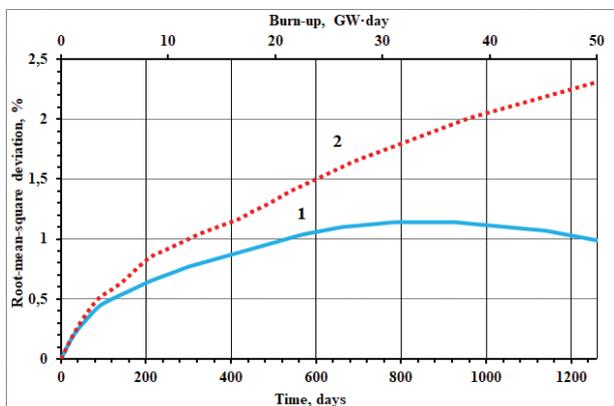


Figure 4. Root-mean-square deviations in the nuclear concentration of  $^{239}\text{Pu}$  caused by uncertainties in the neutron flux (curve 1) and uncertainties in the reaction rates for  $^{238}\text{U}(n,\gamma)$ ,  $^{239}\text{Pu}(n,f)$  and  $^{239}\text{Pu}(n,\gamma)$  (curve 2).

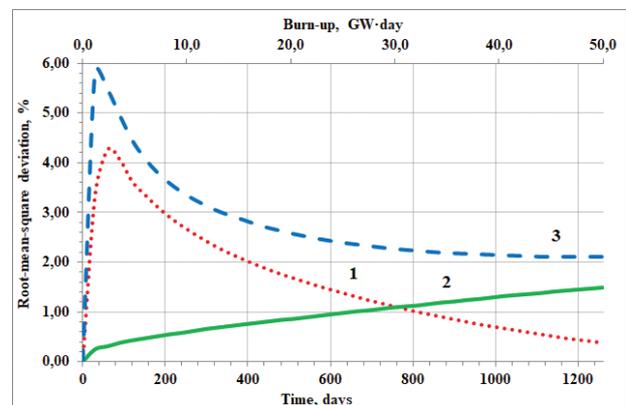


Figure 6. Root-mean-square deviations in the nuclear concentration of  $^{241}\text{Pu}$  caused by uncertainties in the neutron flux (curve 1) and uncertainties in the reaction rates for  $^{241}\text{Pu}(n,f)$  and  $^{241}\text{Pu}(n,\gamma)$  (curve 2) and by uncertainty in the reaction rate for  $^{240}\text{Pu}(n,\gamma)$  (curve 3).

reaction and  $^{240}\text{Pu}$  radiation capture rates for the same reason as for  $^{239}\text{Pu}$ . The uncertainty in the nuclear concentration of  $^{235}\text{U}$  is formed largely by uncertainties in the  $^{235}\text{U}$  fission reaction and radiation capture rates.

An increase or a decrease in the root-mean-square deviation of the nuclear concentrations stems from the change in the behavior of the nuclear concentrations as such. Figs 3, 4 show that an increased rate of the  $^{239}\text{Pu}$  nuclear concentration decay (a diagram bend) leads to its root-mean-square deviation ceasing to grow. A decreased rate of the  $^{241}\text{Pu}$  nuclear concentration buildup leads to its root-mean-square deviations ceasing to decay (see Figs 5, 6).

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## Conclusion

The results of the studies have shown that there is a complex dependence of the root-mean-square deviations in the nuclear concentrations of nuclides caused by uncertainties in the reaction rates and in the neutron flux on burn-up time. As fuel burns up, the behavior of the root-mean-square deviations is not at all times monotonous and depends, for the considered isotope, on the uncertainty source, the reaction rate type, and the precursor nuclei.