VALIDATION OF NUCLIDE KINETICS CODE TRACT FOR THE ASSESSMENT OF SNF AND RW RADIATION CHARACTERISTICS

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The article presents a verification matrix for the TRACT code designed for nuclide kinetics calculations and demonstrates code validation results based on data from reactor experiments. It provides recommendations regarding future experimental SNF studies considered important in terms of assessing the long-term storage and disposal safety of SNF and RW generated from SNF reprocessing.

Keywords: *calculation code, nuclide kinetics, spent nuclear fuel, radiation characteristics, energy release, pressurized water power reactor, verification, validation, reactor, radioactive waste.*

Introduction

Federal Law No. 170-FZ On Atomic Energy Use [1], namely, Article 26, states that the safety assessment of nuclear facilities requires some certified software that should be appropriately validated and verified for this particular purpose. The TRACT software [2], [3] is designed to calculate the radionuclide composition of radioactive waste (RW). Current stage of its development mainly focuses on the radiation characteristics of RW Class 1 and their assessment [4]. The largest inventory of such waste has been accumulated at the PA Mayak site: it involves aluminophosphate glass (APG) resulting from reactor (VVER-440, BN-600, 800, etc.) spent nuclear fuel (SNF) reprocessing and immobilization (vitrification) of the generated liquid radioactive waste (LRW) into the APG waste form. No data on the measured APG radionuclide composition that could be used to validate the TRACT software are available to date. Given the fact that the reprocessed VVER-440 SNF accounts for the largest part of the LRW inventory (over 80%) conditioned into APG upon its solidification, this paper summarizes the software validation results based on the case study of this fuel type.

Since a new VVER-1000 SNF reprocessing plant is going to be commissioned soon (PDC at the MCC site), the article presents the software validation considering the products generated from the reprocessing of this fuel type.

TRACT software for nuclide kinetics calculations

Development of a software product geared to material transmutation and activation calculations focused on some practical issues related to the management of accumulated SNF and RW inventories resulting from the decommissioning of reactor facilities has become an increasingly relevant task. This is basically explained by the existing plans for the decommissioning of RBMK and VVER

Table 1. Isotopes from the SF-Compo experimental database [7]

<table>
<tr><th colspan="4">VVER-1000</th><th colspan="3">VVER-440</th></tr>
<tr><th>NVNPP-5</th><th>KNPP-1</th><th>BaNPP-2</th><th>BaNPP-3</th><th>KoNPP-3</th><th>NVNPP-4</th><th>NVNPP-3</th></tr>
<tr><td colspan="7">^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{243}Am, ^{244}Cm,</td></tr>
<tr><td></td><td colspan="6">^{242}Cm</td></tr>
<tr><td colspan="3">^{234}U</td><td></td><td colspan="3">^{234}U</td></tr>
<tr><td></td><td colspan="5">^{241}Am, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd</td><td></td></tr>
<tr><td colspan="3">^{148}Nd</td><td></td><td colspan="2">^{148}Nd</td><td></td></tr>
<tr><td></td><td colspan="3">^{142}Nd</td><td></td><td>^{142}Nd</td><td></td></tr>
<tr><td></td><td colspan="2">^{236}Pu</td><td></td><td></td><td>^{236}Pu</td><td></td></tr>
<tr><td>^{137}Cs</td><td></td><td></td><td></td><td colspan="2">^{137}Cs</td><td></td></tr>
<tr><td></td><td></td><td></td><td></td><td colspan="2">^{237}Np, ^{155}Gd, ^{151}Eu, ^{153}Eu, ^{154}Eu, ^{155}Eu, ^{147}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{133}Cs, ^{134}Cs, ^{135}Cs, ^{109}Ag, ^{105}Pd, ^{108}Pd, ^{101}Ru, ^{95}Mo</td><td></td></tr>
<tr><td></td><td></td><td></td><td></td><td></td><td>^{245}Cm, ^{246}Cm, ^{242m}Am, ^{154}Sm, ^{148}Sm, ^{150}Nd, ^{140}Ce, ^{142}Ce, ^{99}Tc</td><td></td></tr>
<tr><td></td><td></td><td></td><td></td><td>^{144}Ce, ^{103}Rh</td><td></td><td></td></tr>
</table>

reactor units, which require the evaluation of SNF, irradiated structural materials and RW resulting from SNF reprocessing in terms of their radiation characteristics (RC) especially when it comes to the long-term storage and disposal safety assessments.

The experimental data on the measured mass contents of nuclides from VVER-440 and VVER-1000 SNF is currently available solely as regards some limited number of radionuclides (r/n) (transuranic elements, cesium, neodymium, etc.) and short cooling time periods (up to 10—30 years). These data are deemed insufficient to perform a comprehensive SNF and RW RC analysis, in particular, to provide a reliable long-term safety assessment. Basically, acquisition of such data would require the assessment of longer cooling times and additional consideration of the r/n mass contents (^{129}I, ^{36}Cl, ^{79}Se, etc.) [5], [6].

Table 1 lists the isotopes from the experimental database SF-Compo [7], for which their mass content in SNF was measured. Calculations performed in the TRACT software were validated based on the experimental studies of spent fuel assemblies (SFA) removed from the second and the third unit of Balakovo NPP (BaNPP), the first unit of Kalinin NPP (KNPP) with VVER 440 reactor units, the fourth unit of Novovoronezh NPP (NVNPP) with a VVER-1000 reactor unit. Nevertheless, for the rest of the experiments, there is either some missing measurement data on the SFA and their irradiation modes (for example, as regards the SFA removed from NVNPP-3 and NVNPP-5 units) or no measurement data could be found in the open sources. For example, this is the case of SFA removed from the third unit of Kola NPP (KoNPP).

Since it's quite difficult to measure the hard-to-detect radionuclides, various assessment methods are commonly applied, for example, the radionuclide vector method [5]. Nevertheless, computational study is still considered as the key method. For example, an IAEA Report recommends joint implementation of experimental and computational studies to improve the reliability of assessments focused on irradiated materials and their RC [8].

The TRACT software [2], [3] developed by the Nuclear Safety Institute of RAS can be used to simulate the changes in the isotopic composition of materials occurring due to neutron irradiation causing nuclear coupling and radioactive decay of the resulting unstable nuclides. The mathematical model describing nuclide kinetics is a system of first-order linear differential equations with constant coefficients. Numerical calculations were implemented based on the J. Sidell's matrix exponent method [9]. The software has been certified together with the nuclear data libraries (Table 2) viewed as its integral part.

Most well-known software tools can be considered as international TRACT analogues, for example, FISPACT-2001 designed to calculate radionuclide compositions and included into the package of the European Activation System EASY-2001 [10], ACDAM [11], ORIGEN, ALARA, etc. In Russia, the following software tools were developed and certified to solve nuclide kinetics problems under the fast reactor safety assessments: BPSD/V2.1, CARE_03, TARUSA-9, ORIGEN-2, etc.; for pressurized water reactors: TVS-M, MCU-PD, SFUEL(1.0), CHAIN, etc.; for RBMK reactor units: NUCMA; for EGP-6 reactor unit: nuclear calculator EGP-6 [12].

The TRACT software contains nuclear data libraries developed in advance in a required format [2]. Based on these libraries, one can simulate the changes in the radionuclide composition of a material resulting from its irradiation assuming neuron

Table 2. Set of TRACT nuclear data libraries [2]

№	Nuclear data libraries	Contents of the library
1	Microscopic nuclear data on the neutron-isotope cross sections	704 isotopes from ^{1}H to ^{257}Fm in the neutron energy range of up to 20 MeV
2	Radiation characteristics of a radioactive nuclei	~3,500 radioisotopes from ^{3}H to ^{258}Fm
3	Yields of products during actinide fission by neutrons	44 actinides from ^{227}Th to ^{256}Fm - data on forced fission by neutrons
4	Yields of products during spontaneous actinide fission	15 actinides from ^{232}Th to ^{256}Fm
5	Yields and energies of alpha particles during the radioactive decay	Isotopes from ^{145}Pm to ^{257}Fm with an alpha decay channel
6	Cross sections (α, n) of reactions on light elements with their energy threshold not exceeding the maximum alpha particle energy of 7 MeV	10 elements from Li to Si and their 17 isotopes

energies of up to 20 MeV. These calculations consider all open channels of neutron reactions, including the new nuclei generated in the ground and metastable states, and all the decay channels, the constants for which can be found in the radiation data libraries integrated into the software [2].

Experimental and computational SNF and RW studies selected for the TRACT validation purposes

Some experimental and computational SNF and RW studies providing most complete information required for the computational modeling were selected to verify the TRACT software tool designed to assess the radiation characteristics of SNF, irradiated structural materials and RW resulting from SNF reprocessing, especially considering their long-term storage and disposal times.

The TRACT verification matrix primarily involved the computational SNF and RW studies that are currently considered as most well-known. The handbook [13] can be considered a classic data source providing data on such VVER SNF RCs as activity, integral heat release and its components (alpha, beta, and gamma) that was used in this study to demonstrate the reliability of the calculated SNF RC. Safety Guide RB-093-20 [14] provided the recommended data on the radiation and thermal characteristics of VVER-440, 1000, 1200 SNF.

The second part of the verification matrix involved some experimental studies exploring the irradiated materials, SNF and RW in terms of their RC. [15] presents a most well-known benchmark, namely, the experiment that measured the heat release of fission products upon target nuclei 239Pu, 233U and 235U irradiation with thermal neutrons. This experiment and its results are commonly used in the international benchmark tests [16], [17] to validate the performed calculations.

At present time, lots of experimental studies focused on VVER-440 and VVER-1000 spent fuel burnups discussed in Russian literature sources provide insufficiently detailed descriptions of the experiments, in particular, as regards the SFA irradiation modes (its duration and SFA distribution in different core areas due to the shuffling taking place along the process), SFA cooling times, etc., as compared to those required for the computational modeling purposes, as well as insufficient information on the accuracy of the obtained measurement results. [18] provides a most complete description of an experiment exploring VVER-440 SNF. Under this research implemented under the ISTC 2670r project, the mass yields were measured for 4 uranium isotopes, 11 actinides and 32 fission products in SFA samples cut from the fourth NVNPP power unit. A most informative experimental study on VVER-1000 SNF burnup was chosen for validation purposes: under this experiment, the mass content of isotopes in samples cut from SFAs of KNPP and BalNPP with VVER-1000 reactor units was measured [19]. Compared to the previous experiments, the data were presented for a smaller number of isotopes (3 uranium isotopes, 12 actinides and 5 fission products).

Verification matrix

TRACT software verification and validation was based on experimental and computational studies that have been widely known among the scientific community and actively applied to solve various test problems. These studies could provide most complete information required for the computational modeling. Several test problems were selected. Table 3 presents the verification matrix involving the selected test problems.

Verification and validation results

Test problems 1–2

The handbook [13] provides data on the RC of SNF from VVER-440 and VVER-1000 reactor units. Calculations for test problem 1 and their results were

Table 3. TRACT verification matrix for calculations focused on SNF and RW radionuclide compositions and their RC

№	Purpose of the computation study/ experiment	Reactor unit	Material
1	To calculate the activity and α-, β-, γ-energy release (based on the data from the handbook* [13])	VVER-440 (30 MW day/kgU)	Uranium dioxide, 3.6 % enrichment
2		VVER-1000 (40 MW day/kgU)	Uranium dioxide, 4.4 % enrichment
3	To calculate the residual heat (based on data from the RB-093-20 guideline [14])	VVER-440 (30 MW day/kgU)	Uranium dioxide, 3.6—4.87 % enrichment
4		VVER-1000 (40 MW day/kgU)	Uranium dioxide, 3—4.95 % enrichment
5		VVER-1200 (50 MW day/kgU)	Uranium dioxide, 3.3—4.95 % enrichment
6	To measure the residual heat from the fission products [15—17]	Omega West (OWR)	^{239}Pu, ^{233}U, ^{235}U
7	To measure the mass yields of actinides and fission products (identification of the nuclide composition) [18, 19]	VVER-440	Uranium dioxide, 3.6—4.3 % enrichment
8		VVER-1000	Uranium dioxide, 4.3 % enrichment

*handbook on the radiation characteristics of irradiated nuclear fuel / Kolobashkin V. M., Rubtsov P. M., Ruzhanskiy P. A., Sidorenko V. D. [13] (hereinafter referred to as the handbook)

presented earlier in another paper [4]. As regards test problem 2, Figures 1—3 present reference data and the calculated RC (activity, energy release and its components, specific release of gamma radiation) for the VVER-1000 SNF with 4.4 % initial enrichment in ^{235}U and a burnup of up to 40.48 MW day/kgU depending on the exposure times.

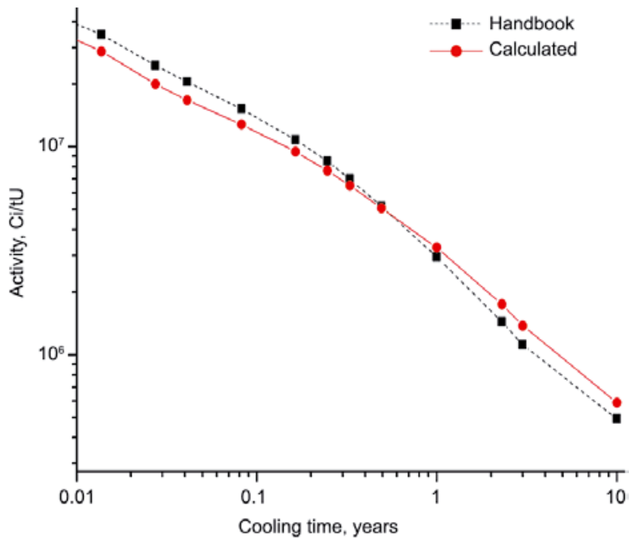

Figure 2. Dependence between VVER-1000 SNF activity and the cooling time

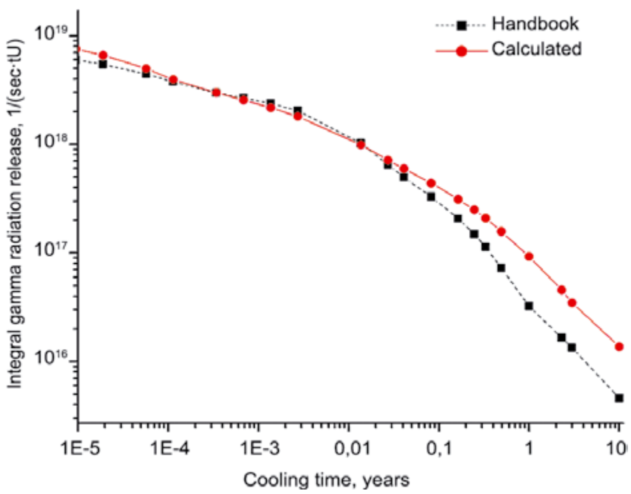

Figure 3. Time dependence for the integral gamma radiation release from VVER-1000 SNF

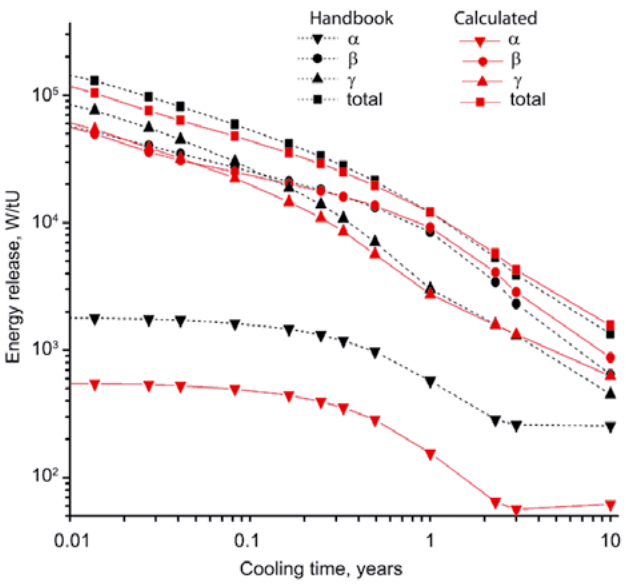

Figure 1. Dependence between the energy release from VVER-1000 SNF and its components and the cooling time

Sets of specific activity and energy release levels calculated for different spent fuel cooling times correlate with the data (at the corresponding time intervals) presented in the handbook [13]. Test calculations focused on the accumulation of fission products and actinides in VVER-1000 SNF showed that the difference between the calculated and reference data on minor actinides lies in the range of (20—70) %. Therefore, the errors in the nuclear data on minor actinides were basically due to the constant libraries applied. For this reason, when these are taken from various libraries of evaluated nuclear data, significant discrepancies may be observed in the calculated results for the accumulated minor actinide masses [20]. Hence, different values may be obtained for the alpha energy release.

Test problems 3—5

RB-093-20 [14] provides data on the radiation and thermal characteristics of VVER-440, 1000, 1200 SNF. For comparison purposes, these were calculated assuming various average initial fuel enrichment by ^{235}U, fuel burnups of VVER-440, VVER-1000 and VVER-1200 SNF and cooling times ranging from 1 to 20 years. Figure 4 presents data on the residual heat release from VVER-440 SNF with ^{235}U fuel enrichment of 4.38 % and a burnup of 30 GW·day/tU calculated using the TRACT software and provides their comparison against relevant levels given in RB-093-20 [14].

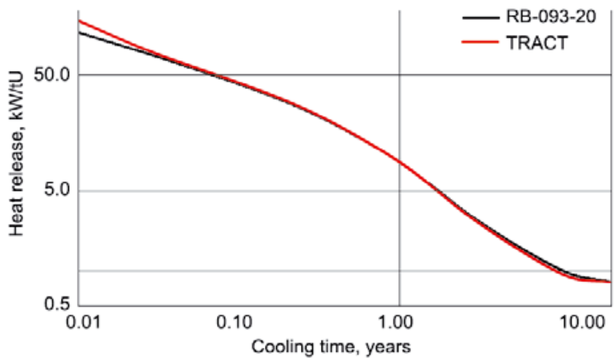

Figure 4. Dependence between the residual heat release of VVER-440 SNF with 4.38 % fuel enrichment by ^{235}U and a burnup of 30 GW·days/tU and the cooling time

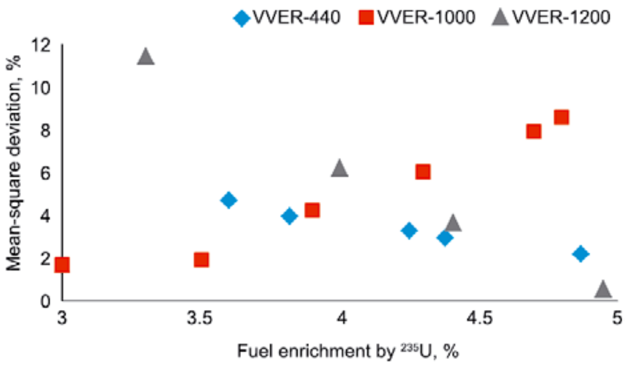

Figure 5. Mean-square deviations of calculated residual SNF heat release from the levels indicated in RB-093-20 [14] depending on the VVER fuel enrichment

Figure 5 shows the mean-square deviations between a set of SNF residual heat calculation results obtained for the test problems 3—5 and the heat release indicators presented in RB-093-20 [14] considering different levels of average initial fuel enrichments with ^{235}U for VVER-440 at a burnup depth of 30 GW·day/tU, for VVER-1000 at burnup depth of 40 GW·day/tU and for VVER-1200 at a burnup depth of 50 GW·day/tU and cooling times ranging from 1 to 20 years.

To compare the values calculated using the TRACT software and the reference or the experimental ones, the deviation (in %) and the mean-square deviation σ of the cumulative results (also in %) were calculated based on the following expressions:

$$\varepsilon = (F_{calc} - F_{mes})/F_{calc} \cdot 100\,\%,$$
$$\sigma_{\varepsilon} = \sqrt{\frac{\sum_{i}^{N} \varepsilon_i^2}{N}}. \quad (1)$$

Mean-square deviations of the calculated residual heat released by VVER-440, VVER-1000 and VVER-1200 SNF from the data presented in RB-093-20 did not exceed 12 %.

Test problem 6

The results of calculations performed under the test problem 6 from the verification matrix were published in [21]. The results obtained showed acceptable agreement between the calculated and the experimental data on the energy release assuming short-time irradiation of thin 235U and 239Pu targets, in which the neutron flux attenuation was considered negligible. Therefore, the algorithm standing behind the developed nuclide kinetics code and the nuclear data libraries on fission product showing relevant decay product yields and the radiation parameters included into the software package could be considered reliable.

The results of this study generally agree with the experimental data within the limits of the measurement errors. The only exception is the energy release calculated for the 233U sample, requiring further refinement of constant libraries considering this isotope.

Test problem 7

The calculation results were published in a conference report [22]. Figures 6 and 7 show the deviations between the calculated mass content of actinides and fission products in sample 57 cut from

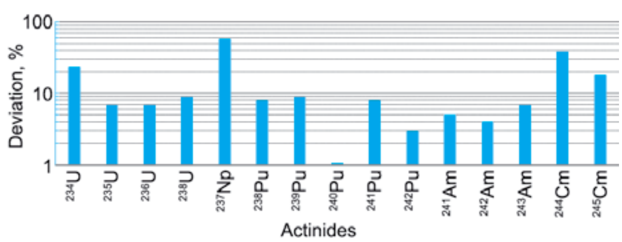

Figure 6. Deviations between the calculated accumulation of actinides in sample 57 cut from VVER-440 SFAs at a fuel burnup level of 36 GW·day/tU and the experimental data

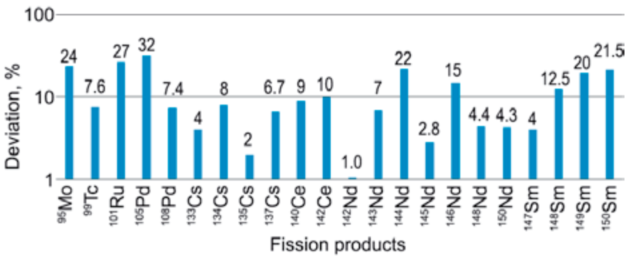

Figure 7. Deviations between the calculated accumulation of fission products in sample 57 from VVER-440 SFA with a fuel burnup of 36 GW day/tU and the experimental data

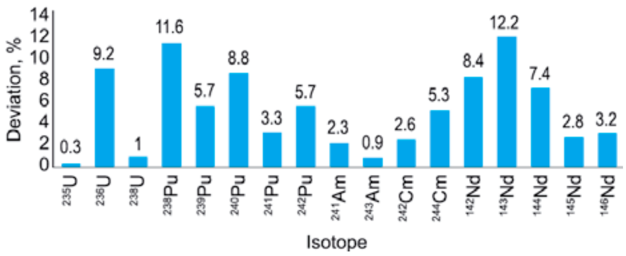

Figure 8. Deviations between the calculated content of actinides and fission products in sample 581 and the experimental data

the VVER-440 reactor SFA with a fuel burnup level of 36 GW·day/tU and the experimental data.

The deviations between the calculated and the measured data for most actinides amount to less than 10 %. For ^{234}U, the accumulation of which amounts to ~100 g/tU_{in}, the deviation accounts for some 24 %. For ^{237}Np and ^{244}Cm it amounts to up to 60 %. It should be noted that such deviations may be also triggered by certain inaccuracies in the experimental results. As noted in [18], for sample 57, the content of ^{237}Np was measured with an error of 39 %, and the one of ^{244}Cm with an error of 33 %. Considering almost the entire burnup region, the data calculated for ^{238}U were found to exceed the measurement results by up to 10 %, which may be due to the underestimated radiative neutron capture cross section assumed according to the applied library data.

Basically, the yields of most fission products are known with an accuracy of ~20 %. The data presented on their accumulation is characterized with quite large discrepancies, mainly as it comes to stable and long-lived fission products with low yields (for example, ^{105}Pd, ^{95}Mo, ^{101}Ru isotopes). This situation may be driven by errors in the experimental data: such error may amount to several tens of a percent. Generally, the comparison between the calculated and the experimental data showed that the considered physical processes could be adequately modelled.

Test problem 8

Relevant calculation results were presented in a conference report [22]. As compared with the data on VVER-440, VVER-1000 measurements involved a smaller number of nuclides. Figure 8 shows the deviations between the calculated content of actinides and fission products from VVER-1000 SFA and the experimental data.

For the ^{236}U isotope, the calculated indicators turned out to be underestimated by up to 10 % for almost the entire burnup region considered. For neodymium isotopes, the deviations did not exceed 2—3 % except for ^{142}Nd and ^{144}Nd with relevant deviations amounting to 11 % and 18 %, respectively.

Cross-verification

For the cross-verification purposes, VVER-440 SNF calculations were performed using the FISPACT program from the EASY-2010 software package [10]. To compare these calculation results with those obtained in the TRACT program, the problem was set similarly to the test problem 1. Figures 9 and 10 present fluctuations in the activity and energy release (alpha, beta, gamma, total) levels depending on a cooling period of up to 1,000,000 years.

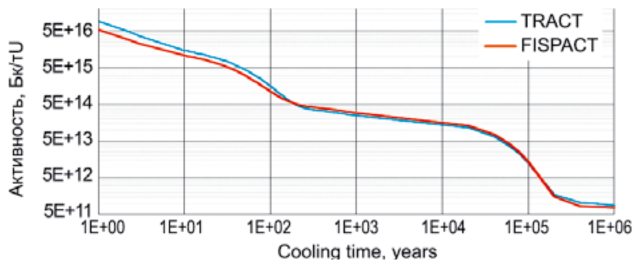

Figure 9. Dependence between the integrated VVER-440 SNF activity and the cooling time of up to 1,000,000 years

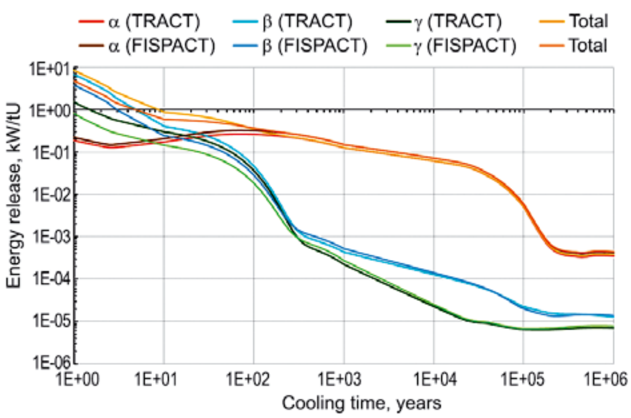

Figure 10. Dependence between VVER-440 SNF energy release (alpha-, beta-, gamma- and total) due to the radioactive decay and the cooling time of up to 1,000,000 years

The fact that the calculations performed using the FISPACT and TRACT software yielded quite similar calculated levels of activities and energy releases shown in Figures 9 and 10 indicated that the TRACT code could be used to calculate the radiation characteristics of SNF from VVER reactor units considering long cooling times of up to 1,000,000 years under the long-term safety assessments.

Conclusion

The article presents validation and verification results for the TRACT software used to evaluate the radionuclide compositions of SNF from VVER-type reactor units, which are required to assess the radionuclide compositions of vitrified RW (APG) resulting from the SNF reprocessing.

Experimental and reference data providing a description sufficient for the computational modeling purposes were analyzed and selected. It was shown that no measurement information was available regarding the content of some radionuclides (^{129}I, ^{36}Cl, ^{79}Se, etc.) important for the long-term safety assessment. Therefore, some experimental research was required to study the radionuclide SNF composition with an account taken of the above isotopes.

Validation and cross-verification based on the calculated RC and the nuclide SNF composition implemented under this study have demonstrated that relevant physical processes could be adequately modelled and such studies could be considered reliable.

The measured mass contents of nuclides in SNF revealed a deviation of up to 10 % between the calculated and the measured data for most actinides. The yields of most fission products were identified with an accuracy of ~ 20 %.

The comparison between the measured data and the calculations based on the FISPACT software demonstrated that the TRACT code could be used in the long-term safety assessments to calculate the RC of spent fuel from VVER reactor units considering a cooling time of up to 1,000,000 years.

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