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CAPABILITIES OF TRACT CODE TO RESOLVE THE PROBLEMS OF RADIOACTIVE WASTE AND SPENT NUCLEAR FUEL CHARACTERIZATION

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The paper presents the calculation capabilities of the nuclide kinetics code TRACT, its validation on the basis of test experiments on the heat release of fission products. Radiation characteristics of VVER-440 reactor spent fuel were analyzed for cooling times up to 10 000 years.

Keywords: *isotope kinetics, code TRACT, validation, decay heat, spent fuel, radioactive waste, gamma-radiation, VVER-440.*

Introduction

Elementary and isotopic composition of materials changes under irradiation due to nuclear reactions and radioactive decay. Development of software analytical systems supporting research on materials transmutation and activation is a fairly resource-intensive and complex task, and has always received considerable attention. The relevance of this task has risen considerably over the last 10-15 years due to higher requirements presented to the accuracy of SNF and RW parameters calculation for the purposes of their long-term storage and disposal [1]. These tasks require calculation of long decay chains and assessing the change in characteristics (activity, energy release, absorbed dose, etc.) of SNF and RW.

One of the relevant tasks is characterization of 1 class RW planned for disposal at the deep disposal facility “Nizhnekansk massif”.

Currently most of 1 class RW generated in RW reprocessing is accumulated at RT-1 plant located at FSUE “PA “Mayak”.

Due to the fact that there was no practical need to identify the radionuclide composition of RW generated in SNF reprocessing, there is a lack of information and considerable indeterminacy of data on the content of a number of key radionuclides in this RW [2]. Minimization of the indeterminacies is one of the key elements of disposal facility long-term safety case. RW characterization for the purposes of safety case has not been performed before, while measurement of radionuclide composition by non-destructive monitoring methods is not possible due to evident reasons. Resolution of this problem does not seem to be possible without application of specialized calculation systems and setting up special conditions for measurements.

TRACT (Transmutation and ACTivation) nuclide kinetics calculation program is being developed for resolution of this problem and other similar tasks. The current paper describes validation studies for the current version of the program TRACT/2018 and gives examples of its testing.

Mathematical model of nuclide kinetics code and development of nuclear data libraries

Nuclide kinetics program TRACT/2018 calculates the change of concentration for material nuclei with account for nuclear transmutations caused by neutron irradiation and radioactive decay. Neutron

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energy may range from thermal to 20 MeV. All open channels of neutron reactions are taken into account in this neutron energy field. The number of channels for some nuclides may reach 18 with account for yield of nuclei in ground and metastable states. Number of metastable states may be up to four. Material may also include non-fissile and fissile elements ranging from hydrogen to fermium (from Z = 1 to Z = 100). All decay channels known from the library of radiation data are taken into account for radioactive decay of radionuclides generated. The output of the neutron kinetics code is the new composition of the irradiated material for a specific time moment of irradiation or storage. The following sets of radiation characteristics are used to assess the properties of the obtained material: activity, spectra of neutron and gamma-radiation, energy release and gas accumulation (hydrogen, helium, tritium) due to nuclear reactions.

Mathematical model of nuclide kinetics is a system of linear first order differential equations with constant coefficients. Isotopic composition of materials changes in process of neutron irradiation due to nuclear reactions and radioactive decay of generated unstable nuclides. The number of nuclei $N_i(t)$ of radionuclides in calculation of nuclide composition of neutron irradiated materials is determined out of a system of differential equations:

$$dN_i(t)/dt = -N_i \cdot (\lambda_i + \sigma_i \cdot \Phi(t)) + \\ + \sum_{j \neq i} (\lambda_{ij} + \sigma_{ij} \cdot \Phi(t)) \cdot N_j + S_i,$$

where $N_i(t)$ — number of radioactive nuclei of i-th nuclide at time moment t;
$\Phi(t)$ — neutron flux, $1/(\mathrm{cm}^2 \cdot \mathrm{s})$;
λ_i — decay constant of i-th nuclide, 1/s;
λ_{ij} — constant of nuclide j decay to nuclide i, 1/s;
σ_i — complete nuclear reactions cross-section at i-th nuclide, cm^2;
σ_{ij} — complete nuclear reactions cross-section at j-th nuclide, leading to yield of i-th nuclide cm^2;
S_i — source of i-th nuclide generation per fission channel, determined as

$$S_i = \sum_k \sigma_k^f \cdot \Phi(t) \cdot N_k \cdot Y_{ik}.$$

Here: N_k — number of fissile nuclei of k-th nuclide;
σ_{kf} — fission reaction cross-section for actinide k;
Y_{ik} — nuclide i due to fission reaction of actinide k.

Calculation of material activation and transmutation in neutron irradiation need to be supported by nuclear data on microscopic interaction cross-sections of neutrons with isotopes of irradiated materials, radiation parameters of radioactive nuclei and fission products yields in actinide fissions by neutrons and gamma-quanta.

After the completion of neutron irradiation, the material, for example SNF, itself becomes a source of the following types of radiation:

- neutrons of spontaneous fission of accumulated actinides and fuel elements;
- alpha-particles generated in radioactive decay of minor actinides. Energy of such alpha-particles does not exceed 7 MeV;
- gamma-quanta generated in radioactive decay of unstable isotopes, mainly fission products, and in spontaneous fission; energy range of such gamma-quanta is up to 12 MeV;
- neutrons of (α, n) reaction taking place at light elements;
- neutrons of (α, n) and (γ, f) reactions taking place at fissile isotopes;

Taking these processes into account requires data on fission products yields in spontaneous fission of actinides, cross-sections of (α, n) reactions at isotopes with energy thresholds below maximum alpha-particle energy of 7 MeV and cross-sections of (α, n) and (α, f) reactions taking place at fissile isotopes.

Nuclear-physical data libraries need to be compiled for all the required data.

Compilation of nuclear data libraries

Neutron cross-sections library

One of the most significant problems to be resolved in development of the code is development of an up-to-date library of nuclear activation data for cross-sections of neutrons interaction with isotopes. These data shall be used to support calculation of activation and change of elementary composition of materials in neutron fields of various nuclear reactors with neutron spectra below 20 MeV. Neutron cross-sections library shall include elements ranging from hydrogen (Z = 1) to fermium (Z = 100), which cover the whole range of both fissile and non-fissile materials.

All neutron reactions possible in the neutron energy range up to 20 MeV shall be described for these nuclides. In addition to stable isotopes, the library shall include nuclear data on radioactive isotopes formed in process of elements transmutations.

The European library EAF-2010 [3] and the Russian library BROND-3/A [4] offer the most information on nuclear activation cross-sections in the neutron energy range up to 20 MeV and represent the best available level of knowledge on assessed nuclear neutron cross-sections. They include elements ranging from hydrogen to fermium (from Z = 1 to Z = 100). The total number of isotopes included in the BROND-3/A library is 704. About 11 000 various neutron reactions possible in the neutron energy range up to 20 MeV are described. In addition to stable isotopes, the library BROND-3/A includes nuclear data on radioactive isotopes formed in process of elements transmutations. Full set of neutron cross-sections for reactions with energy thresholds up to 20 MeV is given for each of the isotopes. This set includes such nuclear reaction channels as: (n, 2n), (n, p), (n, d), (n, t), (n, ^{3}He), (n, α), (n, np), (n, nd), (n, nt), (n, 2p), (n, $p\alpha$), (n, $n\alpha$), (n, γ) and a number of more complex ones of type (n, $n2\alpha$) for the light nuclei range. EAF-2010 library provides data for neutron energy range up to 60 MeV. Both libraries use endf-6 [5] format recommended by the International nuclear data committee as a unified standard format for presentation of assessed nuclear data. A joint start-up activation cross-sections library with preliminary name of TRACT/ACT was formed based on these libraries.

Library of radiation data of radioactive nuclei

The second most important library for the task of calculation of materials activation is the library of radiation parameters of radioactive nuclei formed in nuclear reactions.

Existing libraries include decay data for ~ 3800 radioactive isotopes ranging from ^{3}H to ^{257}Fm, which include all isotopes that may be generated in nuclear reactions. Libraries of isotopic parameters developed in the framework of ENDF/B-VII.1 [9] and JEFF-3.3 [11] projects can be regarded as the most complete ones taking into account both the data of latest experiments, and advanced methods of their analysis. Library TRACT/DEC was compiled based on these libraries and was included in the nuclear database for TRACT/2018 program.

Library of assessed fission products yields nuclear data

Fission products yields are input data of great significance for calculation of SNF radiation parameters. Engineering calculations in the field of reactor physics use close to a thousand values of fission products yields, and thus require available nuclear databases in the form of organized files with a coordinated format structure. Reliability of nuclear installation calculations depends on the quality and reliability of data. New versions of national libraries on fission products yields are currently available, including TENDL-2015 [10], BROND, CENDL-3.1, JEFF-3.1, JENDL-4.0 and ENDF/B-VII.1.

The performed analysis of national libraries of assessed neutron nuclear data on fission products yields has led to development of a compiled library of fission products yields comprising of two parts:

- for 44 actinides ranging from ^{227}Th to ^{256}Fm the data are given for neutron-induced fission;
- for 15 actinides data on fission products yields are given for spontaneous fission.

Library of alpha-particles energies and yields in radioactive decay TRACT/ALPHA

Sets of nuclear data on yield of discrete alpha-particles for radioactive decay of heavy elements were formed based on analysis of the latest available experimental data. The data were prepared using information given in [6] for nuclei range from ^{145}Pm to ^{257}Fm, for the cases where radioactive decay is accompanied by alpha-radiation. Let us note that alpha-radiation energy reaches 6.6 MeV for radon ^{219}Rn. Alpha-particles energy for actinides does not exceed 6.1 MeV.

Library of cross-sections of (α, n) reactions taking place at light elements TRACT/AN

Library of cross-sections of (α, n) reactions taking place at light elements TRACT/AN was based on a compilation drawn up for the project JENDL/AN-2005 [7], and includes data for 17 isotopes: ^{6}Li, ^{7}Li, ^{9}Be, ^{10}B, ^{11}B, ^{12}C, ^{13}C, ^{14}N, ^{15}N, ^{17}O, ^{18}O, ^{19}F, ^{23}Na, ^{27}Al, ^{28}S, ^{29}Si, ^{30}Si. Library JENDL/AN-2005 was prepared in endf-6 format and includes various cross-section s of (α, xn) nuclear reactions in the energy range of α-particles up to 15 MeV.

Library of photonuclear cross-sections at fissile isotopes TRACT/PN

Library of photonuclear data was compiled for fissile isotopes ranging from ^{232}Th to ^{248}Cm. Data for fissile nuclei were taken from the international library of assessed photonuclear data [8] established by an international group of experts managed by IAEA for nuclei ranging from deuterium to plutonium. Some data were taken from the photonuclear data library of projects ENDF/B-VII.1 [9] and TENDL-2015 [11]. Library of cross-sections of (α, n) and (α, f) reactions TRACT/PN was compiled based on these libraries [8—10].

Resolution of test problems for testing the algorithm of energy release calculation in SNF and RW

The accuracy of energy release calculation in SNF and RW depends both on the accuracy of calculation of their nuclide composition for a specific time moment and accuracy of nuclear data. Therefore, there is a need for validation of software used by comparing the calculation results and reference experimental data. The experiments selected for validation should be described in detail, including experimental conditions, results and indeterminacies.

Experiments which could be considered reference for validation of nuclide kinetics code were analyzed, and most informative measurements containing full scope of information required for calculation modeling were selected.

Test problem E1 — measurement of fission products decay heat [12]

Experimental data on heat generation [12], which were used in international comparative tests [13—15] were analyzed as a first step in testing of the developed code. Results of measurements of heat $E(\beta+\gamma)$ generated by thermal neutron fission products of target nuclei ^{239}Pu, ^{233}U and ^{235}U irradiated within a small time interval of $2{\cdot}10^4$ s (~ 5.5 hours) ,were given in [12] for cooling times $T_в$ in the range of 10 to 10^5 s.

Number of fissions in the samples was determined radiochemically out of activity of radioisotopes ^{99}Mo, ^{140}La, ^{147}Nd. For uranium-235 accuracy of measurement of heat generation per a single fission ΔE was within ± 6 %.

Nuclide composition of the initial monoisotopic target nuclei ^{233}U, ^{235}U and ^{239}Pu both during irradiation up to $2{\cdot}10^4$ s (5.5 hours), and in process of cooling was calculated by TRACT/2018 program.

For such relatively small irradiation times the initial composition of the target changes insignificantly and the results of nuclide composition and heat generation calculations are mainly affected by nuclear data on fission cross-sections by thermal neutrons, independent fission products yields, radiation parameters of fission products and actinides.

Nuclear data on fission products yields of ^{233}U, ^{235}U, ^{239}Pu nuclei fission in the calculations were taken from JEFF-3.2 library. This library was selected as the main source of data for TRACT/FPYLD program as it contains the latest most reliable data on fission products yields for ^{233}U, which are used in most calculation studies for uranium-thorium cycle. Analysis of simulation results (Fig. 1—3) leads to a conclusion that for short irradiation times of “thin” ^{235}U and ^{239}Pu targets there was a satisfactory agreement of calculation and experimental data, indicating reliability of the algorithm of the developed nuclide kinetics code and selected nuclear data libraries on fission products yields and radiation parameters. However, the calculation results overestimated heat generation by 15 % for ^{233}U for small irradiation times. This was also noted in the international list of requirements for specification of nuclear data HPRL (Nuclear Data High Priority Request List) [16], which was prepared by international experts in the framework of WPEC (Working Party on International Nuclear Data Evaluation Cooperation) [17]. Efforts on specification of these data are continued both under IAEA leadership (program of data revision for a large group of actinides has been completed in 2017, the assessed data were presented for public use) and in the framework of International nuclear data evaluation cooperation (WPEC).

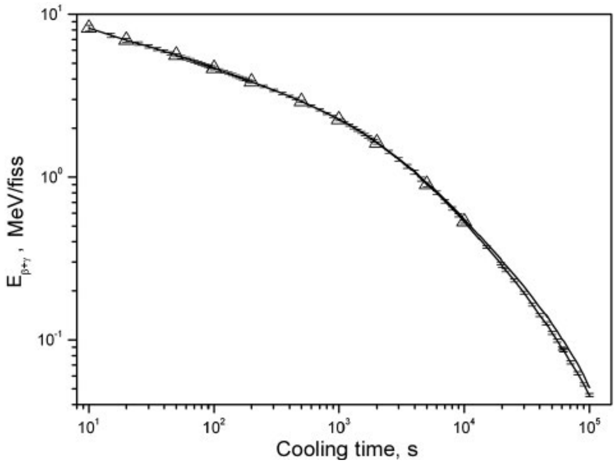

Fig. 1. Experimental and calculation data for heat generation of ^{235}U fission products

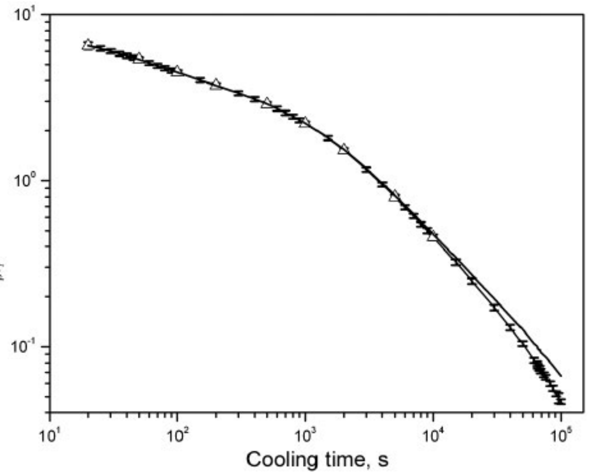

Fig. 2. Experimental and calculation data for heat generation of ^{239}Pu fission products

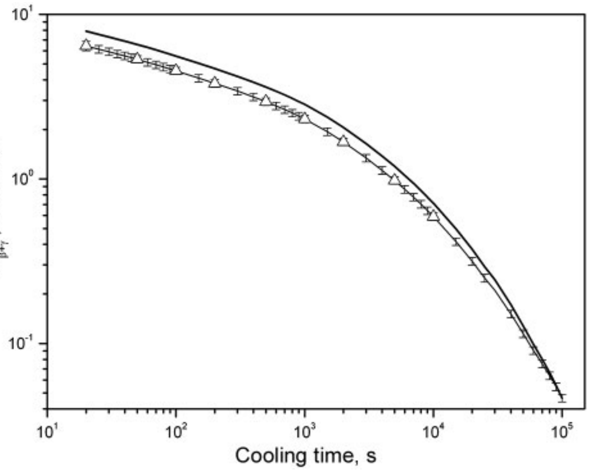

Fig. 3. Experimental and calculation data for heat generation of ^{233}U fission products

Test problem E2 — measurement of fission products energy generation [18—20]

Precision measurements of spectral characteristics of the radiation and residual energy release in irradiated targets containing thorium, uranium and plutonium isotopes were performed in 1980-s at three leading scientific centers (ORNL, LOWELL (USA), YAYOI (Japan) [18—20]).

These measurement results remain the most informative and are used in various international tests both for verification of nuclear data and methods for calculation of nuclide composition and radiation characteristics of nuclear fuel for small irradiation and cooling times.

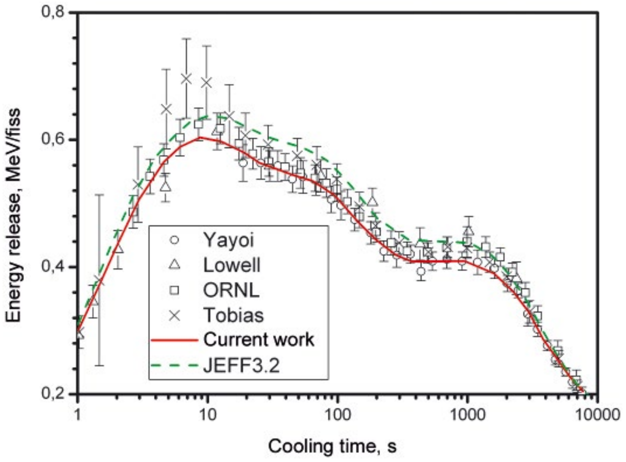

Fig. 4. ^{239}Pu energy release (β-component) as a function of cooling time

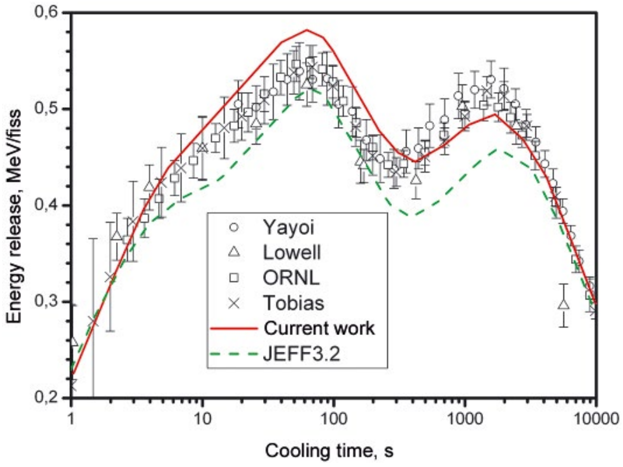

Fig. 5. ^{239}Pu energy release (γ-component) as a function of cooling time

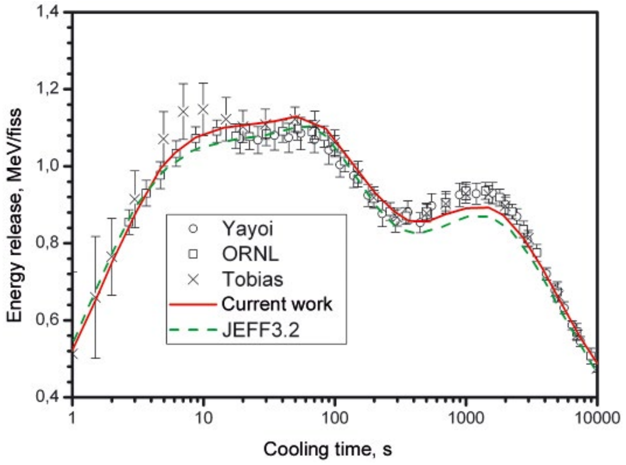

Fig. 6. ^{239}Pu full energy release as a function of cooling time

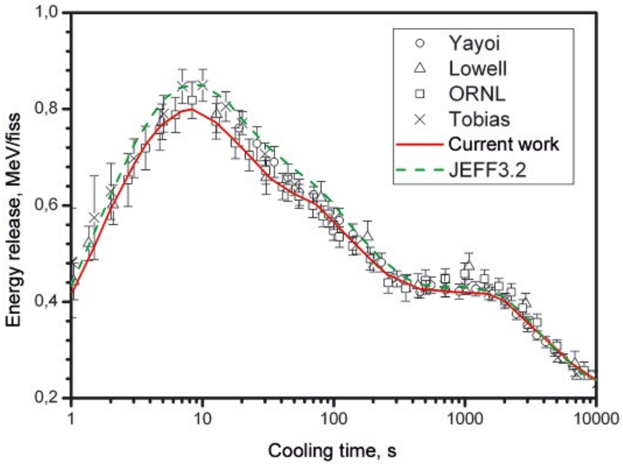

Fig. 7. ^{235}U energy release (β-component) as a function of cooling time

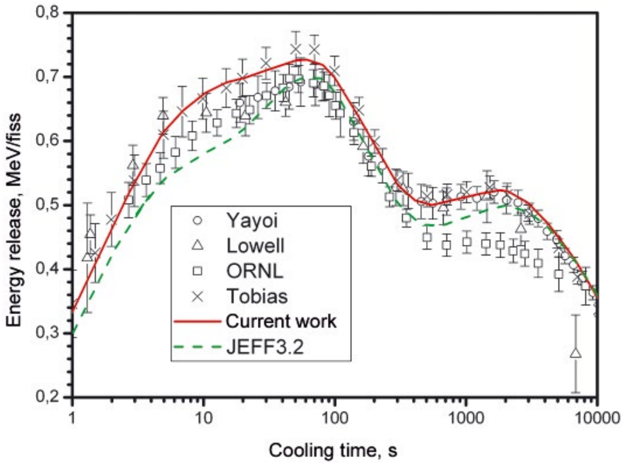

Fig. 8. ^{235}U energy release (γ-component) as a function of cooling time

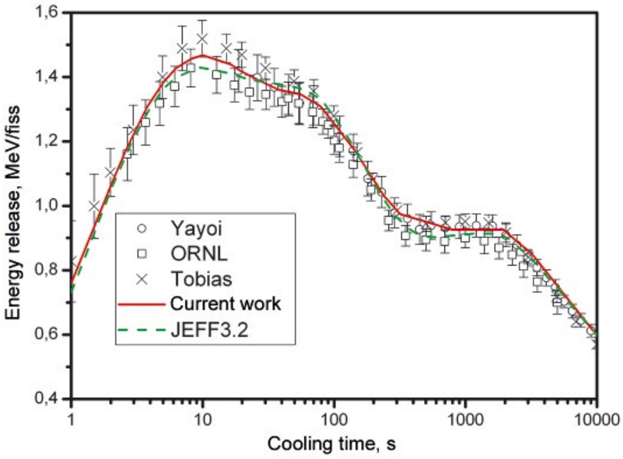

Fig. 9. ^{235}U full energy release as a function of cooling time

Fig. 4—9 show comparison of experimental data of A. Tobias [13], ORNL [18], LOWELL [19], YAYOI [20]. Calculations and comparison were performed for two actinides — ^{235}U and ^{239}Pu. The figures also show calculation data for the same parameters prepared using library JEFF-3.2, which currently includes all the latest data for fission products yields and radiation characteristics (“decay data”).

Assessment of results of calculations using TRACT/2018 program shows that for ^{235}U and ^{239}Pu, both for β- and γ-decay heat components, there was a satisfactory agreement of experimental and calculation data, thus indicating that the nuclear data libraries for these isotopes were selected correctly. These results are confirmed by the analysis of the results of task E1.

Practical testing of the nuclide kinetics program

Radiation parameters of VVER-440 SNF

Calculation of SNF composition formed in operation of VVER-440 reactors, as well as calculation of a number of radiation parameters was performed using the developed nuclide kinetics code. The initial library of neutron activation cross-sections was initially taken in 315-group approximation (TRIPOLI decomposition) and was contracted to a one-group representation with respect to the respective VVER-440 reactor neutron spectrum.

Neutron spectrum at the central part of VVER-440 reactor core in the vicinity of a point at the height of h = 120 cm from the reactor vessel bottom was taken as a representation of neutron field characterizing the core of VVER-440 reactor.

This zone of VVER-440 core is the most energy-intensive and has the highest values of neutron flux in its vicinity.

Uranium dioxide UO_2 fuel (3.6 % enrichment of ^{235}U) was taken for VVER-440. Such fuel is utilized in VVER-440 reactor up to burn-up of 30 GW·day/t UO_2. This variant of fuel burn-up calculation is used for validation of nuclide kinetics programs and was considered in [22].

The calculations of SNF isotopic composition change were performed for storage times up to 10 000 years. It is of interest to compare the obtained results with similar data from reference book [22] (Fig. 10). It should be noted that the compared data are very close. However, if the data in the reference book covered up to 460 fission products ranging from ^{62}Zn to ^{166}Ho, our results give information on approximately 650 fission products in this range of nuclei. Information was also obtained for the isotopes in the range from hydrogen to zinc (about 114 isotopes) and from holmium to thorium.

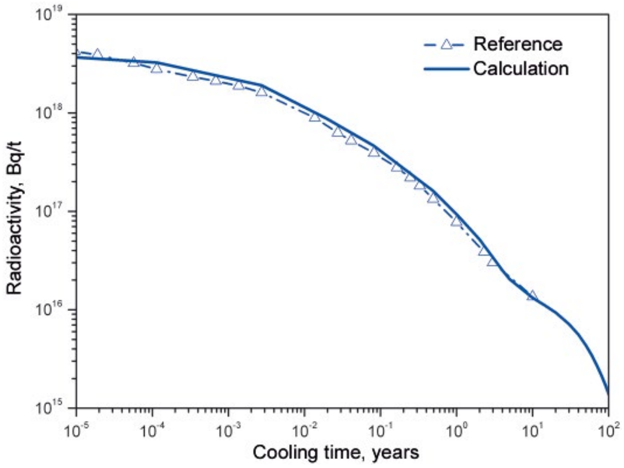

Fig. 10. Comparison of time relations of activity accumulated in SNF for calculation and reference data

An assessment of SNF residual energy release due to emission of γ-quanta, α- and β-particles was performed (Fig. 11).

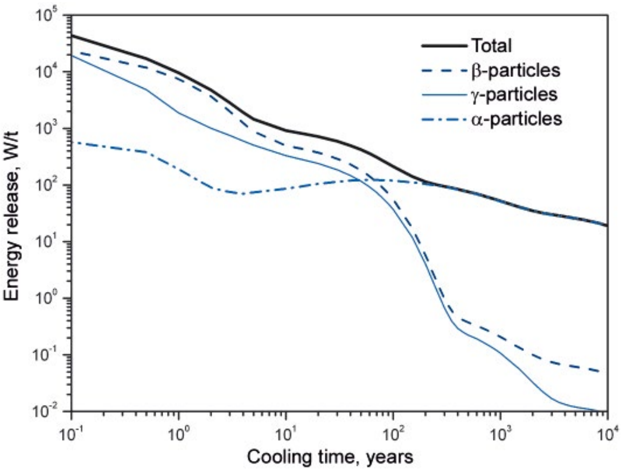

Fig. 11. Total SNF energy release and its components due to beta, gamma, and alpha radiation

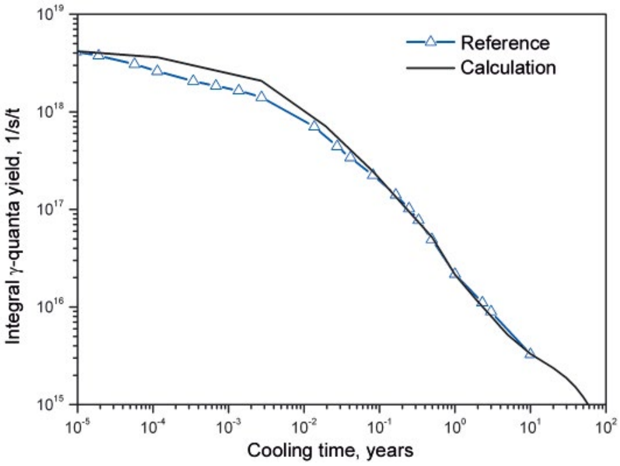

Fig. 12. Comparison of gamma-radiation yields for calculation and reference data

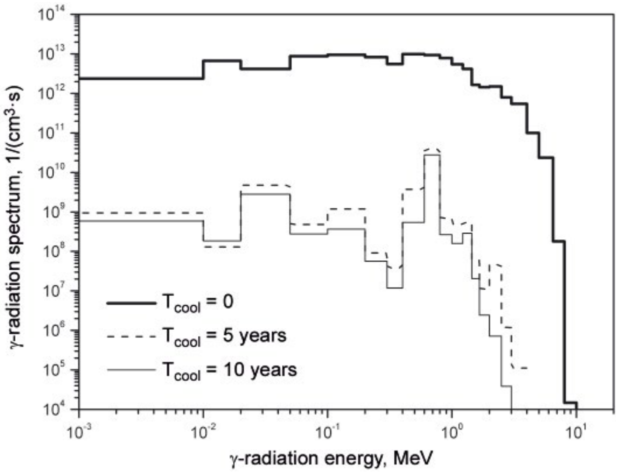

Fig. 13. SNF gamma-radiation energy spectra for different cooling times

"Fresh" SNF contains more short-lived fission products and light isotopes, which emit gamma-radiation in process of decay. Fig. 12 shows comparison of calculation data for γ-radiation yield with similar data taken from [22]. There is an overall good agreement of calculation and test data. It should be noted that the absence of short-lived nuclei in reference data leads to underestimation of γ-yield. Fig. 13 shows calculation data on energy distribution of γ-radiation for various cooling time values.

The results presented provide full picture of SNF composition and radiation parameters.

Radiation parameters of RW produced in reprocessing of VVER-440 SNF

Let us consider radiation parameters of RW produced in reprocessing of VVER-440 SNF. Nuclear fuel fabricated form uranium dioxide UO_2 (3.6 % enrichment of ^{235}U) is used in VVER-440 reactor up to burn-up of 30 GW day/t UO_2, then is stored for 5 years and sent for reprocessing to FSUE "PA "Mayak".

Uranium and plutonium is extracted in process of reprocessing. The resulting RW may contain up to 0.01 % of uranium and up to 0.025 % of plutonium.

According to the calculations, reprocessing of 1 t of SNF generates 154.81 kg of RW. 1.5 to 1.7 tons of vitrified mass is produced in the vitrification process of this quantity of RW, containing 154.81 kg of RW, or 9.1 to 10.32 % of glass mass. We will hence assume that vitrification of 154.81 kg of RW leads to generation of 1600 kg of vitrified mass with a volume of 0.64 m^3 for glass density of 2.5 g/cm^3.

Thus, 1 kg of vitrified RW contains 96.75 g of RW.

Calculations of isotopic composition change were performed for the obtained RW composition for storage times up to 10 000 years. Then residual RW energy release due to beta-particles, alpha-particles and gamma-quanta emission was calculated (Fig. 14). Data are given per 1 kg of initial RW and per 1 kg of vitrified RW.

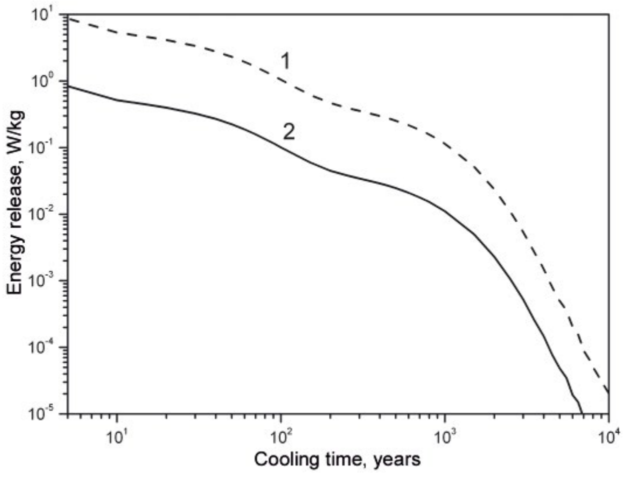

Fig. 14. Energy release for RW (1) and vitrified RW (2)

RW is a source of gamma-radiation. Gamma-radiation spectrum of RW at the time of SNF reprocessing after 5 years of storage, is shown in Fig. 15 compared to the spectra of SNF, uranium and plutonium. It can be seen that the contribution of uranium and plutonium isotopes to gamma-radiation spectrum for the considered SNF is minor. Integral gamma-radiation yield for SNF, uranium and plutonium isotopes is equal to $5.24 \cdot 10^{10}$, $9.72 \cdot 10^5$ and $5.38 \cdot 10^7$ $1/(cm^3 \cdot s)$ respectively.

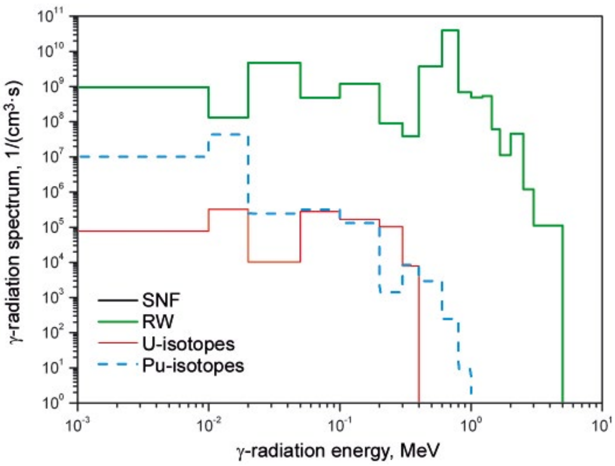

Fig. 15. Energy spectra of γ-radiation of SNF, RW, uranium and plutonium isotopes for storage time of 5 years

Thus, gamma-radiation yield of RW virtually coincides with gamma-radiation of SNF even after extraction of uranium and plutonium from the SNF.

Conclusion

New sets of evaluated nuclear data files for a wide range of nuclei from hydrogen to fermium have been prepared based on critical analysis of the quality of nuclear data in various available data libraries. The following data libraries were developed: nuclear physical data library TRACT/ACT, radiation parameters of radioactive isotopes TRACT/DEC, fission products yields for induced and spontaneous fission TRACT/FPYLD, alpha-particles energies and yields in radioactive decay of actinides TRACT/ALPHA, cross-sections of (α, *n*) reactions at light elements TRACT/AN, cross-sections of (γ, *n*) and (γ, *f*) photonuclear reactions for all available nuclei from thorium-232 to curium-248. A set of test problems was developed for validation of TRACT/2018 program and the prepared nuclear physical data libraries. The set was used to evaluate reliability of calculation of SNF and RW parameters. The calculation data acquired were in good agreement with experimental data (within experimental data error). The only exception was heat generation calculation for ^{233}U. Resolution of this problem requires specification of nuclear data on fission products yields, while this discrepancy does not affect the resolution of practical problems.

The carried out calculations and analysis of results allow the following conclusions:

- algorithm of transmutation and activation calculation of TRACT/2018 works correctly;
- activation data of neutron cross-sections selected for nuclear data library of TRACT/2018 program provided reliable simulation of test problems.

Calculation capabilities were demonstrated and the system was tested by evaluation of SNF composition for VVER-440 reactor and the following parameters of spent fuel: residual energy release, radioactivity, gamma-radiation yields. Gamma-radiation spectra were also assessed for various storage times (0, 5, and 10 years).

It may be concluded that the developed system of programs and nuclear physical data libraries gives adequate description of experimental data and may be utilized as a tool for more complex calculations and analysis of integral reactor and simulated reactor experiments.

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