Analytical Ways of Determining the Activity of Fission Products in the Core of a VVER-1200 Reactor and Their Applications

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Abstract—The activities of fission products in a VVER-1200 reactor thatare of greatest practical importance are calculated analytically. A comparative analysis is performed for the activities of the main fission products (FPs) in VVER-1000 and VVER-1200 reactors. The coefficients of correlation between the complex and easily determined activities of radionuclides in a VVER-1200 reactor are calculated and presented in the form of simple analytical expressions.

DOI: 10.3103/S1062873818100179

INTRODUCTION

Analytical expressions for calculating the specific activities (SAs) of fission products (FPs) in reactor RBMK (high-power pressure tube), VVER-1000, and VVER-440 reactors were obtained in [1–3]. The results were almost equal in accuracy to the values obtained on the basis of numerical simulations. Calculations were performed in the approximation of two and four fissionable nuclides. The results were in good agreement with one another.

In this work, dependences of the rates of burnable isotope fissioning on the duration of the fuel campaign of a VVER-1200 reactor are obtained in the approximation of two fissionable nuclides $(^{235}U$ and ^{239}Pu). The SAs of the FPs most interesting from a practical point of view are calculated in the context of this approximation. The expressions have a simple compact form. Partial contributions to the generated specific activities of FPs due to the fission of 235U and 239Pu are considered.

Correlation ratios for the SAs of pair nuclides in the fuel of VVER-1200 reactor are obtained. A technique for determining at a local level the hard-to-measure activity of nuclides from known easily measurable activities is proposed.

APPROXIMATION OF TWO FISSIONABLE NUCLIDES

It was shown in [1] that the specific mass of 235 U in kg t*–*¹ for VVER-440, VVER-1000, and RBMK-1000 reactors can be approximated to within a few percent (depending on reactor operating time *t*) using the function

$$
m(^{235}U,t) = [m_0(^{235}U) + \mu]e^{-\lambda t} - \mu,
$$
 (1)

where m_0 ⁽²³⁵U) is the initial specific mass of ²³⁵U (kg t⁻¹); μ is a constant; $\lambda \sim 1/\tau$ (years⁻¹), where τ is the reactor campaign and *t* is the time from its start (in years).

It is natural to assume that a function in form (1) can be successfully used for a VVER-1200 reactor.

We can obtain the specific rate of the fissioning of 235 U nuclei if we differentiate (1) with respect to time and multiply it by the corresponding coefficient

$$
P({}^{235}\text{U},t) = P_0 \exp(-\lambda t). \tag{2}
$$

Assuming the mean fission energies of the burnable isotopes are approximately the same, the rate of fission when the reactor is operating at a constant power is

$$
P = P_0 \left[1 - \exp(-\lambda t) \right],\tag{3}
$$

which applies to the remaining fissionable isotopes (mainly $^{239, 241}$ Pu and 238 U). 239 Pu makes the key contribution; the contribution from ²⁴¹Pu for a VVER-1200 reactor is appreciable starting around the middle of a campaign; and the contribution from 241 Pu for VVER-1000 is notable only at the end of a campaign. The total yields of fission products of the 239Pu and ²⁴¹Pu nuclei differ negligibly, so we may assume $P(2^{39,241}Pu, t) \approx P(2^{39}Pu, t)$ without any great error.

The values of the unknown parameters in formula (1) can be found using data from the numerical experiment for a stationary VVER-1200 fuel campaign [4] if we approximate the data with the least-squares method:

$$
m(^{235}U,t) = [m_0(^{235}U) + \mu]e^{-\lambda t} - \mu,
$$
 (1)
$$
m(^{235}U,t) = 55.25 \exp(-0.370t) - 7.29 \text{ (kg t}^{-1}).
$$
 (4)

Expressions (2) and (3) corresponding to burnout dynamics (4) take the form

$$
P({}^{235}\text{U},t) = P_0 \exp(-0.370t), \tag{5}
$$

$$
P({}^{239}\text{Pu},t) = P_0[1 - \exp(-0.370t)], \tag{6}
$$

$$
P_0 = 1.659 \times 10^{18} \text{ fits s}^{-1}.
$$
 (7)

CALCULATING THE SPECIFIC ACTIVITIES OF THE MOST IMPORTANT NUCLIDES IN A VVER-1200 REACTOR

The mass number of most fission products (FPs) of burnable isotopes does not change upon radioactive transformations. This allows us to consider separately the set of FPs with the same mass number, referred to as the isobaric chain.

Most generated FPs have excessive numbers of neutrons. This excess is mainly eliminated due to β [−] decays. With few exceptions, virtually all radiative transformations of fission products are $β$ ⁻ decays.

In addition to β decays, reactions of the radiation capture of neutrons, along with those of type (*n*, 2*n*) and ones even less probable are possible. First, a large cross section of this process is needed in order for these reactions to make an appreciable contribution; second, the nuclide must have a sufficiently long half-life. These processes do not introduce any perceptible error for most isobaric chains. They must be considered along with problems of practical interest only when calculating the activities of 135Xe and 149Sm poisoning nuclei. They can be ignored when calculating the activities of other radionuclides of practical importance.

Let us consider a model of the operating time of fission products that allows for the above assumptions. The rate of change in the number of nuclei for all members of the linear isobaric chain $A_1 \rightarrow A_2 \rightarrow ...$ \rightarrow *A_i* \rightarrow ... \rightarrow *A_n* is described by a system of differential equations whose meaning is completely obvious:

$$
\begin{cases}\n\frac{dA_1(t)}{dt} = \lambda_1 y_1^{\text{ind}} P(t) - \lambda_1 A_1(t) \\
\frac{dA_2(t)}{dt} = \lambda_2 y_2^{\text{ind}} P(t) + \lambda_2 A_1(t) - \lambda_2 A_2(t) \\
\vdots \\
\frac{dA_i(t)}{dt} = \lambda_i y_i^{\text{ind}} P(t) + \lambda_i A_{i-1}(t) - \lambda_i A_i(t) \\
\vdots \\
\frac{dA_n(t)}{dt} = \lambda_n y_n^{\text{ind}} P(t) + \lambda_n A_{n-1}(t) - \lambda_n A_n(t)\n\end{cases} (8)
$$

where *i* is the current number of the linear chain member; λ*ⁱ* is the decay constant of the *i*-th member of the isobaric chain; y_2^{ind} is the absolute independent yield of the *i*-th member of the isobaric chain; and *P*(*t*) is the rate of the given burnable isotope fissioning $(^{235}U, \frac{239}{\text{Pu}}$, and so on).

System (8) is the Bateman problem. Mathematically, this problem has an analytical solution for the rate of fissioning *P*(*t*) of certain types of nuclei (for a constant and exponentially varying *P*(*t*)) in particular).

The rates of burnable isotope fissioning in the approximation of two fissionable nuclides are described by expressions (5) and (6). Fissioning of plutonium nuclei (6) can be conditionally presented in the form of two processes: fissioning with constant velocity P_0 and fissioning with exponentially changing velocity $-P_0$ exp($-0.370t$). The problem of the generation of fission products is reduced to solving system (8) for three different $P(t)$, and the resulting activities are represented by the sum of the solutions:

$$
A_i(X_i, t) = A_i(X_i, {}^{235}U, t)^{\beta}
$$

+ $A_i(X_i, {}^{239}Pu, t) - A_i(X_i, {}^{239}Pu, t)^{\beta}$. (9)

Upper index β means this solution corresponds to *P*(*t*) with an exponential dependence. Analytic expressions for the terms on the right-hand side of (9) were given in [1].

Expression (9) allows us to calculate the specific activities (SAs) of most main fission products, depending on the duration of the fuel campaign.

Nuclides with lifetimes on the order of several hours, days, or years can be of interest in evaluating the SAs in burnup fuel. Nuclides with lifetimes much shorter than the considered time interval can in this case be excluded from consideration by adding their independent yields to the subsequent members of the isobaric chain. System (8) is then usually reduced to 1–2 equations.

Let us find expressions for estimating the dependence of the specific activity of FPs (Bq t*–*¹) on time in a VVER-1200 reactor for nuclides of the greatest practical significance by applying the described approach:

$$
A(^{90}\text{Sr},t)
$$

= (3.48 - 0.43 $e^{-0.370t}$ - 3.05 $e^{-0.0240t}$) \times 10¹⁶, (A1)

 05

$$
A(^{95}Zr,t)
$$

= (7.99 + 3.11 $e^{-0.370t}$ - 11.10 $e^{-3.96t}$) \times 10¹⁶, (A2)

$$
A(^{95}\text{Nb}, t) = (7.99 + 3.28e^{-0.370t} - 24.48e^{-3.96t} + 13.21e^{-7.24t}) \times 10^{16},
$$
 (A3)

 θ ²⁷⁰

$$
A(^{103}\text{Ru},t)
$$

= (11.59 - 6.95 $e^{-0.370t}$ - 4.64 $e^{-6.45t}$) \times 10¹⁶, (A4)

$$
A(^{106}\text{Ru},t)
$$

= (7.21 - 14.42 $e^{-0.370t}$ + 7.21 $e^{-0.677t}$) \times 10¹⁶, (A5)

Fig. 1. Dependence of activities of the most important nuclides on reactor operating time during the campaign.

$$
A(^{131}\text{I},t)
$$

= (6.40 - 1.63 $e^{-0.370t}$ - 4.77 $e^{-31.56t}$) \times 10¹⁶, (A6)

$$
A(^{137}\text{Cs}, t)
$$

= (10.94 + 0.05 $e^{-0.370t}$ - 10.99 $e^{-0.023t}$) \times 10¹⁶, (A7)

 140

$$
A(^{140}\text{Ba}, t)
$$

= (8.87 + 1.49 $e^{-0.370t}$ - 10.36 $e^{-19.85t}$) \times 10¹⁶, (A8)

$$
A(^{141}\text{Ce}, t)
$$

= (8.70 + 1.06 $e^{-0.370t}$ - 9.76 $e^{-7.79t}$) × 10¹⁶, (A9)

$$
A(^{144}\text{Ce},t)
$$

= (6.20 + 5.05 $e^{-0.370t}$ - 11.25 $e^{-0.889t}$) \times 10¹⁶, (A10)

$$
A(^{147}\text{Nd},t)
$$

= (3.32 + 0.42 $e^{-0.370t}$ - 3.74 $e^{-23.06t}$) \times 10¹⁶. (A11)

Graphs of functions A1–A11 are shown in Fig. 1.

COMPARING THE SPECIFIC ACTIVITIES OF THE MOST IMPORTANT NUCLIDES IN THE BURNUP FUEL OF VVER-1200 AND VVER-1000 REACTORS

The chart for determining the SAs of the fission products which are most important from a practical point of view in the burnup fuel of VVER-1000 and VVER-1200 reactors (Fig. 2) can be constructed using the data in [1] and in this work.

There is an approximate equality of activities for most nuclides, except for 106 Ru and 137 Cs. With the former, the cumulative yield of 106 Ru in the fissioning of $235U$ is only 0.41%, while its yield in the fissioning of 239 Pu is 4.19% (a difference of 10 times!), and the amount of fissioned plutonium greatly exceeds the corresponding amount for a VVER-1000 reactor

Fig. 2. Comparative diagram of the activities of the main nuclides in the burnup fuel of VVER-1000 reactors (white bars) and VVER-1200 (shaded bars).

during a VVER-1200 fuel campaign, as was shown earlier. With 137Cs, a VVER-1200 has a 55% longer campaign and operates at a 6.6% higher thermal power. The cumulative yield of 239 Pu relative to 137 Cs is 5.9% higher than the yield relative to 235U. Nuclides with approximately the same activity have similar cumulative yields for uranium and plutonium, as along with relatively short half-lives that allow their saturation during a fuel campaign.

ACTIVITY OF LONG-LIVED γ-EMITTING NUCLIDES IN A VVER-1200 REACTOR

Such γ-emitting fission products as ⁸⁷Rb ($T_{1/2}$ = 4.88×10^{10} years), ⁹³Zr ($T_{1/2} = 1.5 \times 10^{6}$ years), ⁹⁴Nb ($T_{1/2}$ = 2.4 × 10⁴ years), ⁹⁸Tc ($T_{1/2}$ = 4.2 × 10⁶ years), ⁹⁹Tc ($T_{1/2}$ = 2.13 × 10⁵ years), ¹²⁶Sn ($T_{1/2}$ = 1.0×10^5 years), and ¹²⁹I ($T_{1/2} = 1.7 \times 10^7$ years) have long half-lives.

The development of these nuclides during the operation of reactor obviously proceeds linearly, since decaying nuclei can be ignored during a fuel campaign. The activity of these fission products at the end of the fuel campaign can be calculated in the approximation of two fissionable nuclides:

$$
A_X = \lambda_X [y_X^c (2^{35} U) N (2^{35} U) + y_X^c (2^{39} Pu) N (2^{39} Pu)],
$$
 (10)

where $N(X)$ is the number of nuclei of a given nuclide fissioned for a campaign (these can be determined by integrating (5) and (6) over the length of the campaign); y_X^c are cumulative yields in the fissioning of a

Nuclide	$T_{1/2}$, year	λ , year ⁻¹	v^{c} (²³⁵ U)	$y^c(^{239}Pu)$	$A(X)$, Bq t ⁻¹
87Rb	4.9×10^{10}	1.42×10^{-11}	2.58×10^{-2}	1.19×10^{-2}	1.88×10^{6}
^{93}Zr	1.5×10^{6}	4.62×10^{-7}	6.37×10^{-2}	4.11×10^{-2}	1.70×10^{11}
94 Nb	2.4×10^{4}	2.89×10^{-5}	2.47×10^{-9}	1.62×10^{-7}	1.49×10^{7}
98 Tc	4.2×10^{6}	1.65×10^{-7}	8.91×10^{-9}	4.16×10^{-9}	7.59×10^3
99 Tc	2.13×10^5	3.25×10^{-6}	6.14×10^{-2}	6.22×10^{-2}	1.40×10^{12}
126 Sn	1.0×10^5	6.93×10^{-6}	5.61×10^{-4}	1.98×10^{-3}	6.12×10^{10}
129 I	1.7×10^{7}	4.08×10^{-8}	5.39×10^{-3}	1.32×10^{-2}	2.64×10^{9}

Table 1. Specific activities of long-lived γ-emitting nuclides at the end of a VVER-1200 reactor fuel campaign

given fuel nuclide; and λ_X is the decay constant of nuclide *X*.

The results from calculations using (10) are presented in Table 1.

ANALYZING THE PARTIAL CONTRIBUTIONS FROM 235U AND 239Pu BURNABLE ISOTOPES TO THE PRODUCTION OF FPs IN A VVER-1200 REACTOR

The solutions to problem (8) for 235 U and 239 Pu will be considered separately in order to estimate the contributions from individual burnable isotopes to the total production of FPs. The first term (9) will correspond to uranium, and the second and third terms will correspond to plutonium. A long-term fuel campaign of a VVER-1200 reactor is associated with large contributions from 239Pu, especially at the end of the campaign. The graphs of production for the specific activities of FPs are shown in Figs. 3 and 4 for 235U and ²³⁹Pu, respectively.

It should be noted (Fig. 3), that there is very low production of 106Ru SA, which is generated almost completely by plutonium fissioning (Fig. 3).

Fig. 3. Partial contribution of ²³⁵U to the production of the specific activities of FP in a VVER-1200 reactor.

Comparison of the graphs in Figs. 3 and 4 shows that contributions to the activity of burnup fuel for a VVER-1200 reactor are mainly associated with the fissioning of plutonium, and they are almost completely determined by it for individual products of fissioning.

CORRELATION ANALYSIS OF THE SPECIFIC ACTIVITIES OF FPs FOR PLUTONIUM ISOTOPES

Some radionuclides have clear $γ$ -lines, and their activity is measured quite simply (e.g., $95Zr$ with two γ lines: 0.744 MeV, 44% and 0.757 MeV, 55%). Other nuclides may not participate in γ-decays (e.g., 90 Sr), or their γ-lines have low energies with which background radiation is mixed (e.g., 141 Ce γ-line 0.145 MeV, 49%), so their activity is difficult to measure. The specific activities of fission products are interrelated; their ratios can be expressed as [5]

$$
A(X,t) = \xi(t)A(Y,t),\tag{11}
$$

where $\xi(t)$ can be found as the ratio of theoretically calculated specific activities at time *t*, e.g., according to formulas A1–A11. In the same way, we can associate the specific activities of fission products to the specific α-activity of plutonium isotopes, which can be determined from data on their specific masses [4].

Fig. 4. Partial contribution of 239 Pu to the production of the specific activities of FP in a VVER-1200 reactor.

Fuel burnout W is convenient to use in the physics of nuclear reactors when considering different practical problems. Burnout is the energy (MW day) released in one kilogram of fuel (MW day kg−1). Fuel burnout is proportional to the operating time of a reactor when it is operating at a constant power

$$
W = \alpha t. \tag{12}
$$

The physical meaning of proportional coefficient α is the energy (MW day) released in 1 kg of fuel over one year. It is not difficult to determine, based on known constant P_0 : α = 19.4 MW day (year kg)⁻¹.

Expression (12) is clearly valid if it is considered for the entire core, or for a certain volume of fuel composition, where the rate of fissioning is equal to the mean rate throughout the core during a fuel campaign. Since fuel assemblies (FAs) move during a fuel campaign while trying to maintain a uniform release of energy in each fuel assembly, we shall assume that expression (12) is true for any fuel assemblies in a reactor on average during a fuel campaign. We shall solve the problem of this section in the context of this approximation.

We shall conditionally consider that all fuel assemblies in a core burn out on average at the same rate. We shall therefore deal with mean activities in making experimental measurements of activities. Formula (11) must then be rewritten:

$$
\langle A(X,t) \rangle = \langle \xi(t) \rangle \langle A(Y,t) \rangle. \tag{13}
$$

Expression (13) allows one hard-to-measure activity to be calculated from another activity that is easily measured; ξ(*t*) is the corresponding correlation coefficient. In addition, we shall omit the averaging symbols that denote mean values.

When determining activity in an experiment, the accuracy of the formula is better the closer the experimentally determined activity is to the mean value.

In (13), we move from the time parameter to burnout according to (12), omitting the averaging symbols:

$$
A(X,W) = \xi(W)A(Y,W). \tag{14}
$$

Expression (14) shall underlie our calculations of hard-to-measure activities.

THEORETICAL CORRELATION RATIOS OF ACTIVITIES NECESSARY FOR THE ANALYSIS OF EXPERIMENTAL DATA

Let us find the correlation ratios depending on the burnout value for plutonium isotopes, and for the fission products that are most important from a practical viewpoint. The isotopes of plutonium are 239Pu, 240Pu, ²⁴¹Pu, and ²⁴²Pu; the fission products are ⁹⁰Sr, ⁹⁵Zr, 95Nb, 103Ru, 106Ru, 131I, 137Cs, 140Ba, 141Ce, 144Ce, and 147 Nd. As was shown in [5], the ratios of correlations between bound radionuclides have the form of combinations of linear functions and exponentials. We therefore choose functions of this type as approximants. When composing a set of correlation relation-

Fig. 5. Approximation of correlation ratios for nuclide pairs: (a) $A(^{106}Ru,W)/A(^{95}Zr,W)$; (b) $A(^{239}Pu,W)/A(^{95}Zr,W)$.

ships, we strive to choose the pairs for which dependence ξ(*W*) has the simplest form.

As a result, we succeeded in obtaining an approximation of the correlation ratios for most nuclide pairs using simple functions (linear functions and functions in the form $a + be^{-cW}$). These functions are presented in Table 2.

We separately present the approximations of the correlation ratios between the isotopes of plutonium 239 Pu, 240 Pu, 241 Pu, and 242 Pu (Table 3). Examples of approximations for two pairs of nuclides $A(^{106}Ru, W)/A(^{95}Zr, W)$ and $A(^{239}Pu, W)/A(^{95}Zr, W)$ are shown in Fig. 5.

PETROVSKIY et al.

Table 2. Approximation of correlation ratios by analytic functions

Table 3. Correlation ratios between plutonium isotopes

Nuclides ⁹⁵Zr, ¹⁴¹Ce, and ¹⁴⁴Ce are considered to be rigidly bound to the fuel matrix. The ratios between the average experimental activities $A(^{95}Zr)$, $A(^{141}Ce)$, and $A(^{144}Ce)$ must therefore be close to the theoretical one for reactor fuel. They can be used as a criterion for the reliability of experimental data.

CONCLUSIONS

Simple analytical functions that allow us to calculate the activity of VVER-1200 reactor fission products as a function of the length of a fuel campaign were obtained for the most practical of them. The corresponding technique was described. The specific activities of the main fission products in the burnup fuel of VVER-1200 and VVER-1000 reactors were compared.

A procedure for determining the hard-to-measure activity of fission products using ones that are easily determined was proposed on the basis of the correlation relations between the activities of fission products. Correlation ratios were obtained for most pairs of practically important fission products of a VVER-1200 reactor. The expressions had the form of linear functions and functions in the form $a + be^{-cW}$.

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Translated by I. Obrezanova