SOLUTION OF THE EQUATIONS OF NUCLIDE KINETICS

E. F. Seleznev,[1] Yu. Yu. Drobyshev,[1] S. A. Karpov,[1] and I. R. Bukhtiyarov[2]   UDC 621.039.51

Many methods and computational programs are now available for solving burnup equations. Three fundamentally different approaches are combined in a single control program OpenBPS: solution of the nuclide kinetics equation by expansion of a matrix exponential, an iterative method with the possibility of analyzing uncertainties, and a direct analytical solution. The matrix exponential solution is obtained using the modern Chebyshev rational approximation method CRAM. An iterative method taking into account the errors of the initial data and data on decay and cross sections, made possible by an exponential representation of the increment of the nuclide concentration at the BPSE computational step, affords the possibility of analyzing the uncertainty of the nuclear concentration. An analytical computational method based on the use of modified Bateman functions, which is the basis of the ASBE computational code, was developed for the accelerated solution of the burnup equations. Using this program, a solution is obtained more than 100 times faster than the solution speed of programs based on the iterative method with high solution accuracy.

In practice, many methods and programs are available in the practice of solving the burnup equations. Three fundamentally different approaches standout among them: the solution of the nuclide kinetics equation by means of a variety of methods of decomposition of the matrix exponential, an iterative method with the possibility of analyzing uncertainties, and a direct analytical method of solution. All three of these methods of solution were implemented in the all-inclusive code OpenBPS.

Burnup equations. The system of equations of nuclide kinetics is an inhomogeneous linear system of equations

$$\frac{dy_i}{dt} = \sum_{j=1}^{n} a_{ij} y_j + q_i \quad (i = 1, 2, ..., n)$$

with the initial condition

$$y_i(0) = y_{i0} \ (i = 1, 2, ..., n)$$

and an external source

$$q_i \ (i = 1, 2, ..., n)$$

with constant coefficients a_{ij} [1–3]. It possesses a general analytical solution, which is the sum of a particular solution and a general solution of the corresponding homogeneous linear system of equations.

Analytical solution method. An algorithm for obtaining an analytical solution of the system of equations of nuclide kinetics with a full matrix transitions – the first algorithms for solving the burnup problem – was proposed in 1910 [1] with the use of Bateman functions of first kind

$$b_i(t; \lambda_1, \lambda_2, ..., \lambda_i) = \sum_{j=1}^{i} d_j \exp(-\lambda_j t) \tag{1}$$

and second kind

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[1] All-Russia Research Institute for Operation of Nuclear Power Plants (VNIIAES) Moscow, Russia; e-mail: e.f.seleznev@yandex.ru.
[2] National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Moscow, Russia.

$$B_i(t; \lambda_1, \lambda_2, ..., \lambda_i) = \sum_{j=1, j \neq l, m}^{i} d_j A_j - \sum_{j=1, j \neq l, m} d_j A_l + \left(\prod_{j=1}^{i-1} \lambda_j \Bigg/ \prod_{j=1, j \neq l, m}^{i-1} (\lambda_j - \lambda_l) \right) \frac{1}{\lambda_l} [A_l - t \exp(-\lambda_j t)], \tag{2}$$

where

$$A_j = 1 - \exp(-\lambda_j t)/\lambda_j; \quad d_j = 1 \quad (i = j = 1);$$

$$d_j = \prod_{k=1}^{i-1} \lambda_k \Bigg/ \prod_{k=1, k \neq j}^{i} (\lambda_k - \lambda_j) \quad (i \geq 2, j = 1, ..., i). \tag{3}$$

The currently best known program based on this method is DCHAIN [2]. This program shows the exact solutions, including during the first few seconds of the burnup process.

Since some nuclides decay at the same rate, the algorithm of the analytical solution has problems because the difference of the decay rates of nuclides occurs in the denominator. In [2] this problem is solved by representing the ratio of the difference of exponentials with the same exponent, divided by the difference of these exponents, by the duration of the considered time interval, which is correct from the standpoint of mathematics. But this algorithm has not previously considered the indicated problem for the next nuclides in the chain after these two, and for them the solution can no longer be represented by the indicated functions. Moreover, in this algorithm there exists the problem that such a solution is used when cyclic chains are considered.

The solution algorithm for the program ASBE, created for calculating the burnup of actinides and fission products, is based on an analysis of the physical decay process in nuclide chains and, in the absence of singularities, coincides with the solution based on modified Bateman functions (1)–(3).

An analysis of the physical process of the decay of a nuclide in the presence of features associated with the characteristics of the decay rate of other nuclides in the chain affords individual solutions for each nuclide in the chain that are not Bateman functions.

Let the nuclide k have an initial concentration $y^k(0)$ and, in addition, let there be a constant source q^k of its formation. This source affords an increment of the nuclide number k over time τ as $q^k \tau$ without taking into account decay over the time interval $(0, \tau)$. Taking into account the decay of this nuclide over time τ, we obtain its concentration at the time τ (Fig. 1):

$$y^k(\tau) = y^k(0)\exp(-\lambda^k \tau) + q^k[1 - \exp(-\lambda^k \tau)]/\lambda^k.$$

Then the amount of the nuclide k decomposed in the time $(0, \tau)$ is determined as

$$dy_0^k = y^k(0) + q^k \tau - y_0^k(\tau).$$

The increment of the nuclide k is due to the decay of other nuclides into the given nuclide in interval $(0, \tau)$. For the subsequent description of the transformation of nuclides, the increment of each nuclide i, j is simulated over one step in the interval, obtaining an exact solution, taking into account the probability of transitions $(i \rightarrow k)$ and $(j \rightarrow k)$ from the corresponding nuclides (Fig. 2). The increment can also be modeled approximately in terms of the linear representation in the iterative solution (see curves *3, 4* in Fig. 2). As the number of time steps in increases, the increment of the nuclide k will approach the integral describing curve, which is implemented in the programs of the BPS series [3], but the describing curve can be represented as

$$dy^k(t) = dy^k(\tau)[1 - \exp(-\lambda^k t)],$$

where λ is the exponent of the describing curve.

Such a representation should afford a faster solution, which is implemented in the BPSE program as an exponential representation.

Some of the decayed nuclide k can transform into the nuclide l. Then the concentration of the nuclide l for the time τ will be determined as

$$\tilde{y}_1^l(\tau) = y_0^l(\tau) + \sum_{k \neq l} \frac{\lambda^{k \rightarrow l}}{\lambda^k} dy_0^k. \tag{4}$$

To take into account the decay of the nuclei of the nuclide l formed during the time $(0, \tau)$ from the nuclide k, we shall consider the derivative of the decayed nuclide k as

$$[dy_0^k(t)]' = y^k(0)\lambda^k \exp(-\lambda^k\tau) + q^k[1 - \exp(-\lambda^k\tau)], \tag{5}$$

and we rewrite Eq. (4) as

$$y_1^l(\tau) = y_0^l(\tau) + \sum_{k \neq l} \frac{\lambda^{k \to l}}{\lambda^k} \int_0^\tau [dy_0^k(t)]'\, dt. \tag{6}$$

We shall now introduce into the integrand an exponential function to take into account integral to take into account the decay of a nuclide l formed from a nuclide k over time $(0, \tau)$:

$$y_1^l(\tau) = y_0^l(\tau) + \sum_{k \neq l} \frac{\lambda^{k \to l}}{\lambda^k} \int_0^\tau [dy_0^k(t)]' \exp[-\lambda^l(\tau - t)]\, dt. \tag{7}$$

Considering in a similar way the addition of new nuclides to the chain, we obtain a generalized solution of type (1)–(3) with Bateman integral functions of the first and second kind, respectively. The possible difference of their expressions in this case from those presented in (1)–(3) disappears on switching to a representation of the solution from a sum of the difference of exponents with their coefficients to a sum of exponents with the corresponding coefficients (1)–(3). An expression containing representation

$$[\exp(-\lambda^k\tau) - \exp(-\lambda^l\tau)]/(\lambda^l - \lambda^k) \tag{8}$$

is preferable due to possible problems arising for close values of the terms of the difference in the denominator and, together with the presence of a difference of exponentials with these values in the numerator, provides a definite solution even if the denominator is zero.

The problem arising when the decay rates of nuclides k and l are equal will resolve itself in this case. If we use the presented algorithm for obtaining a solution, then for $\lambda^l = \lambda^k$ we obtain from Eq. (8) the value τ.

For the next nuclide in the chain this can be taken into account through Eqs. (5)–(7). After the indicated features, the solution has certain originality, which is also present in the consideration of the subsequent nuclides in the chain. Let us consider two nuclide chains, each with two nuclides, having the same pairwise decay rates – the 3rd and 4th nuclides in the first and 3rd and 5th in the second case (Fig. 3). Bateman's equations will be the solution for nuclides 1 to 3 in the first chain and 1 to 4 in the second chain. For nuclides 3 and 4 of the first chain and 3 and 5 of the second chain, respectively, the difference of the exponentials divided by the difference of their exponents can be replaced in the solution by the length of the interval $(0, \tau)$. For nuclides 5, 6, and 7 of the first chain and 6, 7 of the second chain, the solution on the influence of the decay of nuclides from the 1st to the 3rd in both chains are original, not Bateman functions.

For the problem of residual heat release, i.e., problems without a source and a stable nuclide at the end of the chain, we can write a separate solution:

$$y^i(\tau) = y_0^i(0) + \sum_{n=1}^{i-1} \left(\prod_{j=1,n} \frac{\lambda^{j \to j+1}}{\lambda^j} \right) y^n(0) \sum_{j=1}^{n} \left(\prod_{k=1}^{n} \lambda^k \bigg/ \prod_{k=1, k \neq j}^{n} (\lambda^k - \lambda^j) \right) [1 - \exp(-\lambda^j\tau)].$$

This will be the third integral function for determining the concentration of the corresponding nuclide, although it is a consequence of the first function for a stable nuclide in the chain. The use of such a representation in the program reduces the solution time in the problems of determining the residual heat release.

If the cycle of two nuclides k and l is closed, then, using the presented method of solving (5)–(7), we obtain the concentration of the nuclide k in the form of three terms, the first of which is the initial concentration, the second is the contribution to the concentration of the first from the decay of the second nuclide l to the first k, the third term is the contribution from the decay of the initial concentration, taking into account the external source of the formation of this nuclide into itself through another nuclide of the cycle. Such a definition is no longer represented by Bateman functions of the first and second kind, but is a strictly individual solution. Thus, using the method presented in this work, the solution can be obtained for any cycles.
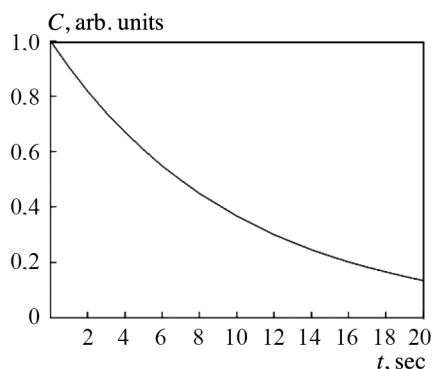
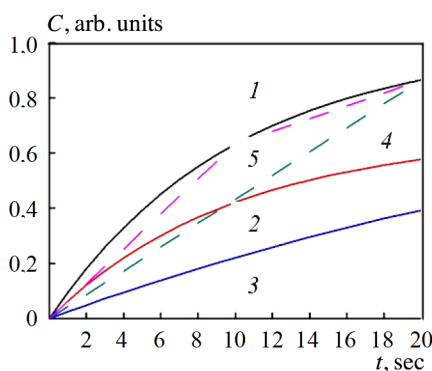Fig. 1. Relative change in concentration of a decaying nuclide over the time.

Fig. 2. Relative change in the increment of the concentration of the nuclide k on the transition of nuclides i and j into it: 1) exp; 2) dy_i; 3) dy_j; 4) $dy - \lin 1$; 5) $dy - \lin 2$.
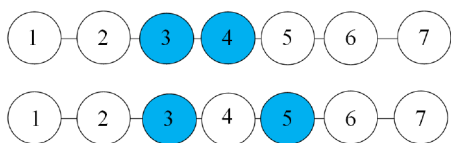

Fig. 3. Chains with nuclides with equal decay rate: 3rd and 4th in the first, 3rd and 5th in the second chain.
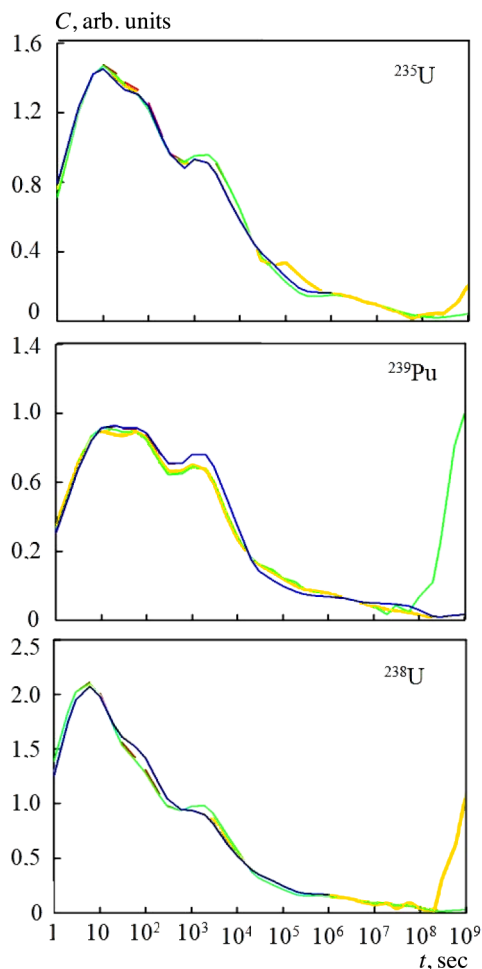

Fig. 4. Time dependence of the residual energy release in a "flash" experiment, calculated using the BPSE (——), CARE (——), ORIGEN (——), and ASBE (——).

Iterative solution method. The essence of the method is to find a solution through knowledge of the derivative, i.e., this is actually Euler's method. This method with linear approximation is implemented in the BPS-series programs, where the solution is determined iteratively with a linear temporal representation of the concentration increment (8):

$$
y_n^k(\tau) = y_{n-1}^k(\tau) + \sum_{m_n \neq k} \frac{\lambda^{m_n \to k}}{\lambda^{m_n}} \frac{1 - \exp(-\lambda^k \tau)}{\lambda^k \tau} \left(1 - \frac{1 - \exp(-\lambda^k \tau)}{\lambda^k \tau}\right)^{n-1} \prod_{j=1}^{n-1} \left(\sum_{m_j \neq k} \frac{\lambda^{m_j \to k}}{\lambda^{m_j}}\right) dy_0^{m_j}.
$$

As already noted, this method with exponential modeling of the nuclide concentration increment is implemented in the BPSE program.

Matrix exponential method of solution. The modern Chebyshev method of rational approximation CRAM was chosen as an algorithm for solving the problem by the matrix exponential method [4–6]. Its features are that the accuracy of the solution weakly depends on the choice of the time step size, the solution time is a linear function of the dimension of the problem, the method is inapplicable for a rapid exponential increase in power, and it is possible to obtain negative concentration values for rapidly decaying nuclides.

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Control program. The open-source program OpenBPS was developed as a control program, which presents the user with a large number of tools for working with input data and when choosing a solution method. The code is based on programs with the initial code in the C++ and Python programming languages and an open-source nuclear data library. The program provides a flexible and user-friendly interface with a choice of any of the listed solution methods and has unlimited possibilities of application at all stages of the fuel cycle – from fabrication and installation into a reactor to assessing the level of activity and residual heat release during reprocessing and conservation of used material.

The results of using the developed programs for calculating the residual energy release on well-known experiments of the "flash" type associated with short-term irradiation for 1 sec with ^{235}U, ^{239}Pu by thermal neutrons and ^{238}U by neutrons with a fast-reactor spectrum are presented in Fig. 4 [6]. The results of calculations using the well-known programs ORIGEN (matrix exponential method) and CARE (analytical solution using Bateman functions) are presented for comparison [7, 8].

Conclusion. The first version of a universal complex of programs for solving the equations of nuclide kinetics was developed, containing the following:

OpenBPS control code with a large number of tools for working with input data and choosing a solution method;

CRAM code based on the Chebyshev rational approximation method when searching for a solution through a matrix exponent;

BPSE code, in which the iterative method is upgraded to an exponential representation of the concentration increment;

ASBE code with analytical solution of kinetic equations.

For the algorithm of an analytical solution of the kinetic equations, the problems involved in solving them disappear when the decay rate of the nuclides in the chain is the same and when the chain is looping, an ASBE code with an analytical solution of kinetic equations was developed, the code volume is about 200,000 operators, the solution speed is 100 times higher than in the iterative method.

Calculations of "flash" experiments were encouraging.

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