

## Solution of Nuclide Burnup Equations Using Transition Probabilities

M. S. Raykin\* and A. I. Shlyakhter

Leningrad Nuclear Physics Institute, Gatchina, Leningrad District, 188350 USSR

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**Abstract**—A new approach to the solution of burnup equations is developed that takes into account the dependence of the reaction constants on time as well as nonlinear and feedback effects. With the help of the transition probabilities for the simplified problem, the burnup differential equation is reduced to the equivalent integral equation, which is solved by iterations. The solution is made easy to understand with the help of diagrams constructed following the suggested rules. It is strictly proved that any nuclide transmutation network can be broken into independent depletion chains if the burnup equations are linear in concentrations. The theory is illustrated by examples of the time dependence of reaction constants.

### I. INTRODUCTION

Problems related to analysis of isotope transmutations are encountered in many areas of nuclear physics and its applications, ranging from star evolution studies<sup>1</sup> and muon catalysis kinetics<sup>2</sup> to optimization of radioactive nuclide production<sup>3</sup> and calculation of the decay heat from nuclear fuel.<sup>4</sup> A detailed network of all possible transmutations may often be very complicated. However, if reaction constants do not depend on the nuclide concentrations, the transmutation network can be broken up into independent linear chains in each of which, if there is no feedback, the isotope concentrations are described by a set of equations:

$$\frac{dx_i}{dt} = s_{i-1}x_{i-1} - \lambda_i x_i, \quad (1)$$

where

$x_i$  = concentration of the  $i$ 'th nuclide

$\lambda_i$  = its total depletion constant

$s_{i-1}$  = constant of its formation from the predecessor ( $s_0 = 0$ ).

If  $s_i$  and  $\lambda_i$  are constant, Eq. (1) can be solved using the simple analytical formula derived by Bateman<sup>5</sup> as early as 1910. Under initial conditions of  $x_i(0) = \tilde{x}_i \delta_{i1}$ ,  $\delta_{ik}$  being the Kronecker delta, the solution can be written as

$$x_n(t) = \tilde{x}_1 \prod_{i=1}^{n-1} (s_i t) \cdot D_n(\lambda_1 t, \dots, \lambda_n t). \quad (2)$$

Here,  $D_n(y_1, \dots, y_n)$  is the depletion function introduced by Siewers<sup>6</sup>:

$$D_n(y_1, \dots, y_n) = \sum_{i=1}^n \frac{\exp(-y_i)}{\prod_{\substack{j=1 \\ j \neq i}}^n (y_j - y_i)}. \quad (3)$$

\*Present address: Boston University, Physics Department, 590 Commonwealth Ave., Boston, Massachusetts 02215.

Properties of these functions were studied in detail in Refs. 7 and 8, in which a number of new represen-

tations are given that are also applicable when the values of some arguments are close or identical (see Appendix B). The depletion functions were applied to analyze the criteria conventionally used in identifying the important chains and in eliminating short-lived nuclei, as well as to obtain simple formulas for preliminary estimates of the nuclide concentrations and the sensitivity of the results to the values of the constants. In these studies,  $s_i$  and  $\lambda_i$  were assumed to be constant and the feedback absent.

Making allowance for variable reaction constants and for the feedback effect, Eq. (1) can be written as

$$\frac{dx_i}{dt} = s_{i-1}x_{i-1} - \lambda_i x_i + f_i(t, x_1, \dots, x_n) \quad (4)$$

Depending on the explicit form of the functions  $f_i$  in Eq. (4), account is taken of the variation of the reaction rates with time [if  $f_i$  has the form  $f_i = x_{i-1}\varphi_{i-1}(t) - x_i\varphi_i(t)$ ], the feedback effect (if for some  $i$ ,  $f_i$  depends linearly on  $x_{i+1}, \dots, x_n$ ), and the nonlinear effects (e.g., the blocking of the resonance absorption of neutrons). Note that with nonlinear effects, different linear chains are not generally independent. If the reaction constants are known time functions, the solution of Eq. (4) can be presented as a multiple integral, explicitly calculated in simplest cases only. To take into account the feedback, one must know eigenvalues of the nontriangular transition matrix, which have to be computed numerically. Thus in the general case, Eq. (4) cannot be solved analytically.

This study is aimed at developing a new approach to burnup equations that makes it possible to take into account the dependence of the reaction constants on time and nuclide concentrations, as well as the feedback effects.<sup>9</sup> It is based on application of transition probabilities for the unperturbed problem, i.e., the problem with constant reaction rates and no feedback.

Introduced in Sec. II is the transition probability that has the sense of the propagator for the unperturbed problem. With its help, Eq. (1), with given initial conditions, is reduced to an equivalent system of integral equations that is solved by iterations. A diagram method is presented in Sec. III similar to the Feynman diagram technique and permitting an analytical expression for each term of a perturbation theory series when the functions  $f_i(t, x_1, \dots, x_n)$  are polynomials of the concentrations  $x_1, \dots, x_n$ . The subject of Sec. IV is the linear problems, namely, the case of the reaction constants being only time dependent and the feedback case. First-order corrections to the burnup equation solutions for the sudden perturbation and linear time dependence of the reaction constants are given in Sec. V. Appendix A gives proof that any nuclide transmutation network described by the linear burnup equations can be decomposed into a set of independent chains. Appendix B gives basic relationships for depletion functions.

## II. TRANSITION PROBABILITY AND INTEGRAL EQUATION FOR NUCLIDE CONCENTRATIONS<sup>a</sup>

The original set [Eq. (1)] has the following matrix form:

$$\frac{dx}{dt} = \hat{A}x \quad (5)$$

where  $x$  is the concentration vector and  $\hat{A}$  is the reaction rate matrix. We introduce transition probability matrix  $\hat{P}(t)$  in which the element  $P_{ij}(t)$  is the concentration of the  $i$ 'th nuclide at time  $t$  on condition that initially only the  $j$ 'th nuclide with a unity concentration was present, i.e.,

$$P_{ij}(t) = x_i(t)|_{x_k(0)=\delta_{kj}} \quad (6)$$

Thus,  $P_{ij}(t)$  satisfies the equation

$$\frac{dP_{ij}(t)}{dt} = A_{ik}P_{kj}(t) \quad (7)$$

In systems of linear differential equations, matrix  $\mathbf{P}(t)$  is called the matricant of Eq. (5) (Refs. 10 and 11). Equation (7) yields the relationship

$$P_{ik}(t - t_1) \cdot P_{kj}(t_1) = P_{ij}(t) \quad , \quad 0 \leq t_1 \leq t \quad (8)$$

In the case of a successive chain, matrix  $\mathbf{A}$  is two-diagonal ( $A_{ll} = -\lambda_l, A_{l,l-1} = s_{l-1}, l = 1, \dots, n$ ), and the explicit expression for  $P_{ij}(t)$  can be obtained with the help of probabilistic considerations. Indeed, the transition probability is yielded by

$$P_{ij}(t) = \int_0^t dt_{i-1} \int_0^{t_{i-1}} dt_{i-2} \dots \int_0^{t_{j+1}} dt_j \times \prod_{l=j}^{i-1} \left\{ s_l(t_l) \exp \left[ - \int_{t_{l-1}}^{t_l} \lambda_l(\xi) d\xi \right] \right\} \times \exp \left[ - \int_{t_{i-1}}^t \lambda_i(\xi) d\xi \right] \quad (9)$$

Here,  $t_l$  is the instant of transition of the  $l$ 'th nuclide into the  $(l + 1)$ 'th one. The factor

$$s_l(t_l) dt_l \cdot \exp \left[ - \int_{t_{l-1}}^{t_l} \lambda_l(\xi) d\xi \right]$$

is the probability of this transition occurring over the interval from  $t_l$  to  $t_l + dt_l$ , while the last exponent expresses the probability that the  $i$ 'th nuclide does not pass to the next one over the interval from  $t_{i-1}$  to  $t$ . At constant values of  $\lambda_l$  and  $s_l$ , we obtain

<sup>a</sup>To facilitate the notation, it is assumed in Secs. II, III, and IV that the summation from 1 to  $n$  is taken with respect to twice repeated indices.

$$P_{ij}(t) = \int_{\tau_j \dots \tau_{i-1} \geq 0} \dots \int \theta(t - \tau_j - \dots - \tau_{i-1}) \times \prod_{l=j}^{i-1} [s_l d\tau_l \exp(-\lambda_l \tau_l)] \times \exp[-\lambda_i(t - \tau_j - \dots - \tau_{i-1})], \quad (10)$$

where  $\theta(x) = 0$  if  $x \leq 0$ ,  $\theta(x) = 1$  if  $x > 0$ , and  $\tau_l = t_l - t_{l-1}$  is the "waiting time" in the  $l$ 'th nuclide. With the presentation in Eq. (B.3) used for the depletion functions, Eq. (10) yields

$$P_{ij}(t) = \prod_{l=j}^{i-1} (s_l t) D_{i-j+1}(\lambda_j t, \dots, \lambda_i t). \quad (11)$$

The probabilistic interpretation makes it clear that when matrix  $\hat{\mathbf{A}}$  is not two diagonal the total probability  $P_{ij}(t)$  is given by the sum of terms of the form of Eq. (11) over all the transition network paths from the  $j$ 'th to the  $i$ 'th nuclide. With no feedback, the total number of transition paths is finite, and the total transmutation network can be split into a set of a finite number of independent linear chains.<sup>12-14</sup>

Thus, the solution of the set of burnup equations [Eq. (5)] with triangular matrix  $\hat{\mathbf{A}}$  is represented as a finite sum of solutions for the sets of the form in Eq. (1) with two-diagonal matrices made up from  $A_{ik}$  elements. With the feedback present, the splitting is performed similarly, but the number of chains here is infinite (see Sec. IV), and the breakup algorithm must be strictly proved. This representation of the solution can be shown to be valid (see Appendix A) not only for burnup equations but also for an arbitrary system of linear homogeneous first-order differential equations (for example, when the elements of matrix  $\hat{\mathbf{A}}$  are complex numbers, and the problem does not permit a probabilistic interpretation).

Because Eq. (5) is linear, its solution at arbitrary initial conditions  $\mathbf{x}(0) = \tilde{\mathbf{x}}$  has the form

$$\mathbf{x}(t) = \hat{\mathbf{P}}(t)\tilde{\mathbf{x}}. \quad (12)$$

With allowance made for the dependence of the reaction constants on nuclide concentrations and time, Eq. (5) takes the form

$$\frac{d\mathbf{x}}{dt} = \hat{\mathbf{A}}\mathbf{x} + \mathbf{f}(t, \mathbf{x}), \quad \mathbf{x}(0) = \tilde{\mathbf{x}}, \quad (13)$$

where  $\mathbf{f}(t, \mathbf{x})$  is the general form of the additional term describing the above-mentioned effects. The transition probability has the sense of the propagator; therefore, the set of differential equations [Eq. (13)], together with the initial conditions, can be reduced to an equivalent set of integral equations. In our case, the system has the form

$$\mathbf{x}(t) = \mathbf{x}^{(0)}(t) + \int_0^t dt_1 \hat{\mathbf{P}}(t - t_1) \cdot \mathbf{f}[t_1, \mathbf{x}(t_1)], \quad (14)$$

where  $\mathbf{x}^{(0)}(t)$  is the solution for the "unperturbed" system [Eq. (5)] with the same initial conditions, obtained from Eq. (12). Indeed, by operating on both parts of Eq. (13) by  $d/dt - \hat{\mathbf{A}}$  and by using the relationships in Eqs. (6) and (7), we can see that  $\mathbf{x}(t)$  satisfies Eq. (13). Furthermore, at  $t = 0$  the second term in Eq. (14) disappears, and because  $\mathbf{x}^{(0)}(0) = \tilde{\mathbf{x}}$ , the initial conditions are also met.

Regarding the second term in Eq. (14) as the perturbation, we obtain the solution to the integral equation by successive iterations constructed as follows:

$$\mathbf{x}^{(n)}(t) = \mathbf{x}^{(0)}(t) + \int_0^t dt_1 \hat{\mathbf{P}}(t - t_1) \times \mathbf{f}[t_1, \mathbf{x}^{(n-1)}(t_1)]. \quad (15)$$

The solution of Eq. (13) by the iterations of Eq. (15) is known<sup>11,15</sup>; however, this approach has never been applied to burnup equations in the perturbation theories.

It can be shown by conventional methods that at any finite interval  $[0, T]$  for a uniform convergence of the iterations at  $n \rightarrow \infty$  to the accurate solution  $\mathbf{x}(t)$ , it is sufficient that the partial derivatives of the functions  $f_i$  by the concentrations  $x_j$  be bounded over the entire domain of the function

$$\left| \frac{\partial f_i(t, x_1, \dots, x_n)}{\partial x_j} \right| \leq \Lambda. \quad (16)$$

The condition is satisfied in cases of practical importance. Note, however, that the rate of iteration convergence to the exact value of  $\mathbf{x}(t)$  generally depends on  $t$ ; the number of iterations required to obtain the pre-set accuracy level may increase with larger values of  $t$ .

It can be shown<sup>9</sup> that the adjoint functions used in the perturbation theory for burnup equations<sup>3</sup> are proportional to  $P_{ij}(t - t_1)$ , while the concentration variation  $\mathbf{x}^{(1)}(t) - \mathbf{x}^{(0)}(t)$  coincides with the result of the first iteration [Eq. (15)].

### III. DIAGRAM TECHNIQUES

When the additional terms  $f_i(t, x_1, \dots, x_n)$  are polynomial functions of the concentrations  $x_1, \dots, x_n$ , the iteration solution in Eq. (15) permits a transparent interpretation. For instance, take

$$f_j(t, x_1, \dots, x_n) = U_{jkl}(t)x_k x_l. \quad (17)$$

By substituting this expression into the formulas for the first two iterations and taking into account the relationship of Eq. (12), we obtain

$$x_i^{(1)}(t) = P_{ij}(t)\tilde{x}_j + \int_0^t dt_1 P_{ij}(t - t_1) \times U_{jkl} P_{kp}(t_1)\tilde{x}_p \cdot P_{lq}(t_1)\tilde{x}_q \quad (18)$$

and

$$\begin{aligned}
 x_i^{(2)}(t) &= P_{ij}(t)\tilde{x}_j + \int_0^t dt_1 P_{ij}(t-t_1)U_{jkl}(t_1) \\
 &\times \left[ P_{kk_1}(t_1)\tilde{x}_{k_1} + \int_0^{t_1} dt_2 P_{kk_1}(t_1-t_2) \right. \\
 &\quad \times U_{k_1k_2k_3}(t_2)P_{k_2k_4}(t_2)\tilde{x}_{k_4}P_{k_3k_5}(t_2)\tilde{x}_{k_5} \left. \right] \\
 &\times \left[ P_{ll_1}(t_1)\tilde{x}_{l_1} + \int_0^{t_1} dt_3 P_{ll_1}(t_1-t_3) \right. \\
 &\quad \times U_{l_1l_2l_3}(t_3)P_{l_2l_4}(t_3)\tilde{x}_{l_4}P_{l_3l_5}(t_3)\tilde{x}_{l_5} \left. \right].
 \end{aligned}
 \tag{19}$$

When the brackets are opened, it can be easily seen that the expressions for  $x^{(1)}$  and  $x^{(2)}$  are the sums of the products of transition probabilities  $P_{ij}$ , coefficients  $U_{jkl}$ , and initial concentrations  $x_i$  integrated over intermediate time moments within the indicated limits. This makes it possible to present each term of the analytical solution with the help of diagrams similar to the Feynman diagrams that are widely used in the quantum field theory. Let us formulate the diagram-building rules for a nonlinearity of the Eq. (17) type.

1. Each transition probability  $P_{ij}(t_2 - t_1)$  is shown as a straight line segment running from right to left from "point"  $(j, t_1)$  to point  $(i, t_2)$  ( $t_1 \neq t_2$ ).

2. The end of the line that comes to the extreme right edge of the diagram (which refers to the instant  $t = 0$ ) corresponds to an additional factor  $\tilde{x}_j$ ,  $j$  being the index of this end.

3. The coefficient  $U_{jkl}$  is depicted as a vertex to which two lines come from the right with the end indexes  $k$  and  $l$  and from which a single line departs to the left with the zero index  $j$ . The upper incoming line corresponds to the second index ( $k$ ) and the lower one, to the third index ( $l$ ).

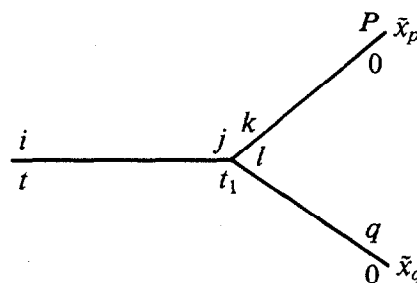
4. The analytical expression corresponding to a given diagram is used to take the summation with respect to all the recurring indexes and the integration over each intermediate moment  $t_k$  from zero to  $t_{k-1}$ . Here,  $t_{k-1}$  is the moment corresponding to the vertex to which comes the line outgoing to the left from the vertex  $t_k$ . The integration is taken from zero to  $t$  over the instant  $t_1$  of the extreme left vertex.

Note that Eq. (14) plays the same role as the Dyson equation in the quantum field theory with respect to the above diagram technique.

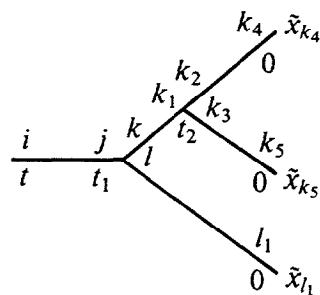
Let us illustrate these rules. The first iteration,

Eq. (18), is contributed to by two iterations only, Eqs. (20a) and (20b). The second iteration, Eq. (19), is contributed to by three diagrams, Eqs. (21), (22), and (23). The respective analytical expressions have the following form:

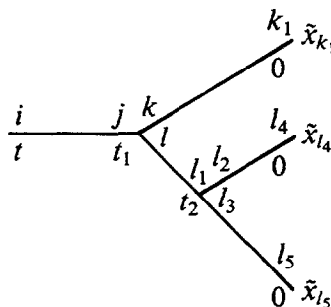
$$\frac{i}{t} \text{---} \frac{j}{0} \tilde{x}_j \quad P_{ij}(t)\tilde{x}_j, \tag{20a}$$



$$\int_0^t dt_1 \cdot P_{ij}(t-t_1)U_{jkl}P_{kp}(t_1)\tilde{x}_pP_{lq}(t_1)\tilde{x}_q, \tag{20b}$$

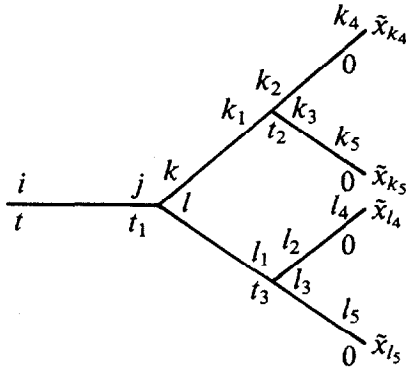


$$\begin{aligned}
 &\int_0^t dt_1 P_{ij}(t-t_1)U_{jkl}(t_1)P_{ll_1}(t_1)\tilde{x}_{l_1} \\
 &\times \int_0^{t_1} dt_2 P_{kk_1}(t_1-t_2)U_{k_1k_2k_3}(t_2) \\
 &\times P_{k_2k_4}(t_2)\tilde{x}_{k_4}P_{k_3k_5}(t_2)\tilde{x}_{k_5}, \tag{21}
 \end{aligned}$$



$$\begin{aligned}
 &\int_0^t dt_1 P_{ij}(t-t_1)U_{jkl}(t_1)P_{kk_1}(t_1)\tilde{x}_{k_1} \\
 &\times \int_0^{t_1} dt_2 P_{ll_1}(t_1-t_2)U_{l_1l_2l_3}(t_2) \\
 &\times P_{l_2l_4}(t_2)\tilde{x}_{l_4}P_{l_3l_5}(t_2)\tilde{x}_{l_5}, \tag{22}
 \end{aligned}$$

and



$$\begin{aligned}
 & \int_0^t dt_1 P_{ij}(t-t_1) U_{jkl}(t_1) \\
 & \times \int_0^{t_1} dt_2 P_{kk_1}(t_1-t_2) U_{k_1 k_2 k_3}(t_2) \\
 & \times P_{k_2 k_4}(t_2) \tilde{x}_{k_4} \cdot P_{k_3 k_5}(t_2) \tilde{x}_{k_5} \\
 & \times \int_0^{t_1} dt_3 P_{ll_1}(t_1-t_3) U_{l_1 l_2 l_3}(t_3) \\
 & \times P_{l_2 l_4}(t_3) \tilde{x}_{l_4} P_{l_3 l_5}(t_3) \tilde{x}_{l_5} . \quad (23)
 \end{aligned}$$

For any iteration, the diagrams are tree shaped, with two lines coming from the right to each vertex and one line going out to the left in conformity with the quadratic form of the nonlinearity [Eq. (17)]. The zero approximation  $\mathbf{x}(t) = \mathbf{x}^{(0)}(t)$  yields the simplest diagram, namely, a line running to the right edge that corresponds to the moment  $t = 0$ . The pattern of each branch is identical to that of the tree; i.e., it either reaches the right edge or splits into two more lines.

Each diagram of the  $m$ 'th order, i.e., one with  $m$  vertexes, represents  $m$  perturbations affecting the formation of the  $i$ 'th nuclide at fixed moments  $t_1, \dots, t_m$ . Their total contribution to the concentration  $x_i(t)$  is given by the respective analytical expression. For example, the first-order diagram, Eq. (20), shows that at time  $t$  when  $x_k(t_1) = P_{kp}(t_1) \tilde{x}_p$ ,  $x_l(t_1) = P_{lq}(t_1) \tilde{x}_q$ , the derivative  $x_j$  changed by the value of the additional term  $f_j(t, x_1, \dots, x_n)$  in the right part of the  $j$ 'th equation of Eq. (4). During the time from  $t_1$  to  $t_1 + dt_1$ , this will change  $x_j$  by  $\Delta x_j(t_1) = dt_1 \Delta x_j(t_1)$ , making  $x_i(t)$  change by

$$\begin{aligned}
 \Delta x_i(t) &= P_{ij}(t-t_1) \cdot \Delta x_j(t_1) \\
 &= P_{ij}(t-t_1) U_{jkl}(t_1) P_{kp}(t_1) \\
 & \times \tilde{x}_p P_{lq}(t_1) \tilde{x}_q dt_1 ,
 \end{aligned}$$

and the integral will sum the contributions from all such events over the range  $0 \leq t_1 \leq t$ .

Comparing the diagrams to the analytical expressions obtained from the above formulas, it can be seen that the diagram series provides solution of the system

[Eq. (14)] with nonlinearity [Eq. (17)]. Indeed, after summing all the diagrams, we obtain

$$\begin{aligned}
 x_i(t) &= P_{ij}(t) \tilde{x}_j + \int_0^t dt_1 P_{ij}(t-t_1) \\
 & \times U_{jkl}(t_1) x_k(t_1) x_l(t_1) , \quad (24)
 \end{aligned}$$

because any diagram other than the simplest one has the shape of a line splitting at point  $t_1$ . These branch lines, in their turn, give rise to two diagram series that provide accurate solutions for  $x_k(t_1)$  and  $x_l(t_1)$ .

Diagrams are similarly constructed for perturbations of the kind

$$f_i(t, x_1, \dots, x_n) = U_{ij_1 \dots j_k}(t) x_{j_1} \dots x_{j_k} \quad (25)$$

with only the third rule modified: Each vertex is approached from the right side by  $k$  lines, the uppermost one corresponding to the index  $j_1$ , the next one to  $j_2$ , and the lowermost one to  $j_k$ . If the perturbation is a added; thus the iteration and the diagram series are powers  $k_l, l = 1, \dots, m$ , the result is contributed to by all the diagrams in which the left-right direction line can branch into any number  $k_1, \dots, k_m$  of lines.

Now compare the two methods of solving the set of integral equations [Eq. (14)], namely, the iterations according to Eq. (15) and the diagram techniques. Any diagram of the  $n$ 'th iteration is a line going to the left from the vertex to which the diagrams of the  $(n-1)$ 'th iteration come from the right; thus, the  $n$ 'th iteration is contributed to by all the diagrams that have no more than  $n$  vertexes on any path from the left to the right edge. Included here are all the diagrams up to the  $n$ 'th order as well as many higher order diagrams. Note that to obtain, at the  $n$ 'th iteration, a solution whose perturbation accuracy would be up to the  $n$ 'th order, it is sufficient to take account, at each preceding iteration, of only those terms whose order  $m$  does not exceed the number of the respective iteration  $i$ . This follows from the fact that the diagram of the order  $m > i$  gives rise, after the  $(i+1)$ 'th iteration, to the diagram of the order  $m' > i+1$ .

Thus, each iteration, starting with the second one, corresponds to the summation of a certain class of diagrams. This obviously adds merits to the iteration approach by making it unnecessary to sort out several diagrams. In addition, when the condition [Eq. (16)] is met, the iteration algorithm is applicable to arbitrary perturbations, while the diagram techniques are only valid with respect to those that can be presented as a sum of terms of the form of Eq. (25). On the other hand, it can often be necessary just to find a first-order correction, or to allow for the feedback effect, and to take into account the reaction rate dependence on time (linear cases). Here both methods are equivalent, while the more comprehensible diagrams prove useful both for writing down respective analytical expressions and for interpreting them.

IV. LINEAR CASE

IV.A. Time Dependence of Reaction Rates

If the additional terms of Eq. (17) are linear with respect to nuclide concentrations, the diagram technique is considerably simplified. Take, for instance,

$$f_i(t_1, x_1, \dots, x_n) = U_{ik}(t)x_k \quad (26)$$

Here, there is only one line coming to each vertex from the right and going from it to the left. At the  $n$ 'th iteration there is only one  $n$ 'th-order diagram added; thus the iteration and the diagram series are identical. The first three diagrams are shown in Eqs. (27), (28), and (29). The respective analytical expressions have the following form:

$$\frac{i}{t} \xrightarrow{\quad} \frac{j}{0} \tilde{x}_j \quad P_{ij}(t) \tilde{x}_j, \quad (27)$$

$$\frac{i}{t} \xrightarrow{l_1} \times_{t_1}^{m_1} \xrightarrow{j} \frac{j}{0} \tilde{x}_j \quad \int_0^t dt_1 P_{i l_1}(t-t_1) U_{l_1 m_1}(t_1) P_{m_1 j}(t_1) \tilde{x}_j, \quad (28)$$

$$\frac{i}{t} \xrightarrow{l_1} \times_{t_1}^{m_1} \xrightarrow{l_2} \times_{t_2}^{m_2} \xrightarrow{j} \frac{j}{0} \tilde{x}_j \quad \int_0^t dt_1 P_{i l_1}(t-t_1) U_{l_1 m_1}(t_1) \times \int_0^{t_1} dt_2 P_{m_1 l_2}(t_1-t_2) \times U_{l_2 m_2}(t_2) P_{m_2 j}(t_2) \tilde{x}_j. \quad (29)$$

If only a slow time variation of the reaction rates following a given law [ $\lambda_i \rightarrow \lambda_i + \mu_i(t)$ ,  $s_i \rightarrow s_i + \nu_i(t)$ ] has to be accounted for, then for the perturbation  $U_{ik}(t)$  we have

$$U_{ik}(t) = \nu_{i-1}(t) \delta_{i-1,k} - \mu_i(t) \delta_{i,k}. \quad (30)$$

As an example, let us consider the transmutation of  $^{237}\text{Np}$  into  $^{238}\text{Pu}$  analyzed in detail in Ref. 16. Neglecting for simplicity the effect of the short-lived  $^{238}\text{Np}$ , we can write the set of equations in the form

$$\dot{x}_1 = -\Sigma_1 \Phi x_1 - \sigma_1 \delta \Phi x_1, \quad x_1(0) = \tilde{x}_1$$

and

$$\dot{x}_2 = \Sigma_1 \Phi x_1 + \sigma_1 \delta \Phi x_1 - \Sigma_2 \Phi x_2 - \sigma_2 \delta \Phi x_2, \quad x_2(0) = 0. \quad (31)$$

Subscripts 1 and 2 refer to  $^{237}\text{Np}$  and  $^{238}\text{Pu}$ , respectively;  $\sigma_i$  is the thermal neutron capture cross section,

and  $\Sigma_i$  is the effective capture cross section ( $\Sigma_i = \sigma_i + \gamma I_i$  where  $\gamma$  is the spectrum hardness and  $I_i$  the resonance integral). The fluxes of resonance and thermal neutrons are assumed to be constant and variable, respectively:  $\Phi(t) = \Phi + \delta \Phi(t)$ , i.e.,  $\nu_1(t) = \sigma_1 \delta \Phi(t)$ ,  $\mu_i = \sigma_i \delta \Phi(t)$  ( $i = 1, 2$ ). Taking into account the diagrams shown with Eqs. (27) and (28), we obtain

$$x_1^{(1)}(t) = x_1^{(0)}(t) - \int_0^t dt_1 \delta \Phi(t_1) \times P_{11}(t-t_1) \sigma_1 x_1^{(0)}(t_1) \quad (32)$$

and

$$x_2^{(1)}(t) = x_2^{(0)}(t) + \int_0^t dt_1 \delta \Phi(t_1) \times [-P_{21}(t-t_1) \sigma_1 x_1^{(0)}(t_1) + P_{22}(t-t_1) \sigma_1 x_1^{(0)}(t_1) - P_{22}(t-t_1) \sigma_2 x_2^{(0)}(t_1)]. \quad (33)$$

By substituting the transition probability equations [Eq. (11)]

$$P_{ii}(t) = \exp(-\Sigma_i \Phi t), \quad i = 1, 2$$

$$P_{12}(t) = \Sigma_1 \frac{\exp(-\Sigma_1 \Phi t) - \exp(-\Sigma_2 \Phi t)}{\Sigma_2 - \Sigma_1}, \quad (34)$$

one obtains the result of Ref. 16.

IV.B. Feedback Case

If the chain allows transitions of a daughter nuclide to the parent state, Bateman's solution [Eq. (2)] is inapplicable because the matrix of Eq. (4) becomes nontriangular. For instance, the radiative neutron capture and the ( $n, 2n$ ) reaction always give rise to a feedback loop. In fissile reactors where the fraction of sufficiently fast neutrons is low, the effects of this loop, as well as of the alpha-decay feedback, are usually small. For this reason, the calculations that use the analytical solution of the burnup equations either neglect these effects<sup>4,12,14</sup> or make allowance for one or two feedback cycles and keep the chain linear by adding fictitious isotopes<sup>17,18</sup> to it.

The perturbation [Eq. (26)] describes the feedback effect on the  $i$ 'th nuclide by the  $k$ 'th nuclide ( $i < k$ ) if  $U_{ik}(t) = \nu(t) > 0$ . Here, the  $l$ 'th-order diagram depicts  $l$  consecutive cycles along the feedback loop that start at the instants  $t_l, \dots, t_1$  when the  $k$ 'th nuclide is transformed into the  $i$ 'th one. Each iteration that follows takes into account still another cycle:

$$x_n^{(l+1)}(t) = x_n^{(0)}(t) + \int_0^t dt_1 P_{ni}(t-t_1) \nu(t_1) x_k^{(l)}(t_1). \quad (35)$$

If only the first nuclide was present at the initial instant, then at constant reaction rates ( $\nu = \text{const}$ ), using Eq. (2),  $x_n(t)$  ( $n \geq i$ ) can be presented as a series:

$$x_n(t) = x_n^{(0)}(t) \left\{ 1 + \sum_{l=1}^{\infty} \left[ (\nu t) \prod_{m=i}^{k-1} (s_m t) \right]^l \times \frac{D_{n+l(k-i+1)}(\lambda_1 t, \dots, \lambda_n t, \{\lambda_i t\}_l \dots \{\lambda_k t\}_l)}{D_n(\lambda_1 t, \dots, \lambda_n t)} \right\}. \quad (36)$$

Here,  $\{\lambda t\}_l$  denotes a sequence of  $l$  coinciding arguments, and the  $l$ 'th term of the sum corresponds to the allowance for  $l$  cycles along the feedback loop yielding a linear chain of  $n + l(k - i + 1)$  nuclides. As demonstrated in Appendix A, the sum over all such chains or "trajectories" gives the exact solution. To compute depletion functions with several identical arguments, one can use Eqs. (B.1), (B.4), and (B.5). A matrix expansion, similar to Eq. (36), was obtained in Ref. 19; however, the recurrent relationships used therein for matrix diagonalization are inapplicable when the burnup rates of some nuclides are identical.

To estimate the terms of Eq. (36), one can use either the inequalities [Eq. (B.7)] or the method proposed in Ref. 20; in many cases it suffices to take into account a single feedback cycle. For simplicity, let us assume that  $s_m = \lambda_m = \lambda_n = \lambda$ ,  $m = 1, \dots, n - 1$ . Then, Eq. (B.10) is reduced to Eq. (B.4) and, taking into account only the term with  $l = 1$  in Eq. (36), we obtain

$$x_n^{(1)}(t) = x_n^{(0)}(t) \cdot \left[ 1 + \frac{\nu}{\lambda} \cdot \frac{(\lambda t)^{k-i+1}}{n \dots (n+k-i)} \right]. \quad (37)$$

Hence, it becomes obvious that if  $\nu \ll 1$ , the feedback loop, which acts as the delay line, is important only at a long irradiation time  $\lambda t > n$  when the concentration of the desired nuclide starts decreasing after passing the peak.

## V. PARTICULAR CASES OF REACTION RATE TIME DEPENDENCE

### V.A. Sudden Perturbation

Analysis of this case makes it possible to find coefficients of the nuclide concentration sensitivity to the accepted values of the constants. Assuming, Eq. (30),  $\nu_k(t) = \nu_k \theta(t)$  and  $\mu_k(t) = \mu_k \theta(t)$ , we obtain in the first order  $x_n^{(1)}(t) = x_n^{(0)}(t) + \delta_1 + \delta_2$ ; the diagrams for  $\delta_1$  and  $\delta_2$  are shown in Eqs. (38a) and (38b) and the respective expressions have the following form:

$$\delta_1 = \sum_{k=1}^n \delta_1^{(k)} = \sum_{k=1}^n \nu_k \int_0^t dt_1 P_{n,k+1}(t-t_1) x_k^{(0)}(t_1) \quad (38a)$$

and

$$\delta_2 = \sum_{k=1}^n \delta_2^{(k)} = \sum_{k=1}^n \mu_k \int_0^t dt_1 P_{n,k}(t-t_1) x_k^{(0)}(t_1). \quad (38b)$$

Hence,

$$\frac{\partial x_n^{(0)}(t)}{\partial s_k} = \frac{x_n^{(0)}(t)}{s_k} \quad (39)$$

and

$$\frac{\partial x_n^{(0)}(t)}{\partial \lambda_k} = -t \prod_{i=1}^{n-1} (s_i t) D_{n+1}(\lambda_1 t, \dots, \lambda_n t, \lambda_k t), \quad (40)$$

which can be easily checked to be identical to the results obtained from the recurrence relationships for the depletion functions.<sup>8</sup>

### V.B. Linear Dependence

Over small time intervals, slowly varying reaction constants can be approximated by a linear dependence. The perturbation-describing terms in Eq. (30) are written as

$$\nu_k(t) = b_k t$$

and

$$\mu_k(t) = a_k t. \quad (41)$$

The analytical expressions of Eqs. (28) and (29) are convolution-type integrals, which makes it easy to use the Laplace transform. The result for the transition probability  $P_{ij}(t)$  is

$$\bar{P}_{ij}(q) = \int_0^{\infty} \exp(-qt) P_{ij}(t) dt = \frac{s_j \dots s_{i-1}}{(q + \lambda_j) \dots (q + \lambda_i)}. \quad (42)$$

By using the correspondence  $t \cdot f(t) \doteq -(\partial/\partial q) \bar{f}(q)$ , the first iteration [for the initial conditions  $x_i(0) = \delta_{i1} \bar{x}_1$ ,  $i = 1, \dots, n$ ] yields

$$x_n^{(1)}(q) = x_n^{(0)}(q) - \bar{x}_1 \sum_{k=1}^{n-1} b_k \bar{P}_{n,k+1}(q) \frac{\partial}{\partial q} \bar{P}_{k,1}(q) + \bar{x}_1 \sum_{k=1}^n a_k \bar{P}_{nk}(q) \frac{\partial}{\partial q} \bar{P}_{k,1}(q), \quad (43)$$

where  $x_n(q)$  is the Laplace transform of the function  $x_n(t)$ . Taking into account Eq. (42) and the identity

$$\frac{\partial}{\partial q} \frac{1}{(q + \lambda_1) \dots (q + \lambda_k)} = \sum_{i=1}^k \frac{\partial}{\partial \lambda_i} \frac{1}{(q + \lambda_1) \dots (q + \lambda_k)}, \quad (44)$$

we find

$$x_n^{(1)}(q) = x_n^{(0)}(q) - \sum_{k=1}^{n-1} b_k \sum_{i=1}^k \frac{\partial x_n^{(0)}(q)}{\partial \lambda_i} - \sum_{k=1}^n a_k \lambda_k \frac{\partial}{\partial \lambda_k} \sum_{i=1}^k \frac{\partial x_n^{(0)}(q)}{\partial \lambda_i (1 + \delta_{ik})} \quad (45)$$

By taking the inverse Laplace transform and using Eq. (40), we obtain

$$x_n^{(1)}(t) = x_n^{(0)}(t) \left[ 1 + \sum_{k=1}^{n-1} (b_k t) \times \sum_{i=1}^k \frac{D_{n+1}(\lambda_1 t, \dots, \lambda_n t, \lambda_i t)}{D_n(\lambda_1 t, \dots, \lambda_n t)} - \sum_{k=1}^n (a_k t) (\lambda_k t) \times \sum_{i=1}^k \frac{D_{n+2}(\lambda_1 t, \dots, \lambda_n t, \lambda_i t, \lambda_k t)}{D_{n+1}(\lambda_1 t, \dots, \lambda_n t, \lambda_i t)} \right] \quad (46)$$

For instance, for a chain with equal burnup rates ( $\lambda_i = \lambda$ ,  $a_i = \mu$ ,  $i = 1, \dots, n$ ;  $b_i = \nu$ ,  $i = 1, \dots, n - 1$ ), with Eq. (B.4) taken into account, we get

$$x_n^{(1)}(t) = x_n^{(0)}(t) \left[ 1 + \frac{n-1}{2} (\nu t) - \frac{(\mu t)(\lambda t)}{2} \right] \quad (47)$$

If  $n = 11$ ,  $\lambda = \mu = 0.01$ , the relative error associated with the approximation of constant reaction rates is 12.5% at  $t = 5$  and -5% at  $t = 12$ .

### VI. SUMMARY

The following conclusions can be drawn:

1. A new approach to nuclide burnup equations is developed that makes it possible to take into account the dependence of the reaction constants on time as well as of the nonlinear and feedback effects. Rules are suggested for constructing diagrams that make it possible in most cases to write easily interpreted analytical solutions. It is strictly proved that any nuclide transmutation network described by linear burnup equations can be decomposed into independent successive chains.

2. The examples considered demonstrate applicability of our approach for developing approximate analytic solutions. Although the clear trend in practical nuclear reactor computations has been in the opposite direction, such formulas remain valid for simple cases, estimates, and teaching.

### APPENDIX A

#### DECOMPOSITION OF THE NUCLIDE NETWORK INTO INDEPENDENT CHAINS

It is generally considered self-evident that any nuclide network described by linear burnup equations can be decomposed into a number of independent successive chains. However, this statement has never been strictly proved. To give such proof, it is demonstrated here that the solution to a set of first-order linear differential equations with an arbitrary (e.g., complex) matrix can be represented as the sum of known solutions to sets of particular form with two-diagonal matrices.

Consider the problem

$$\frac{dx}{dt} = \hat{\mathbf{A}}\mathbf{x}(t), \quad \mathbf{x}(0) = \bar{\mathbf{x}}, \quad (A.1)$$

where  $\hat{\mathbf{A}}$  is any  $n$ 'th order square matrix assumed for simplicity to be so far independent of  $t$ . Thus,

$$\mathbf{x}(t) = \exp(\hat{\mathbf{A}}t)\bar{\mathbf{x}} = \lim_{m \rightarrow \infty} (\hat{\mathbf{I}} + \hat{\mathbf{A}}dt)^m \bar{\mathbf{x}}, \quad (A.2)$$

where  $dt = t/m$ , and  $\hat{\mathbf{I}}$  is the unity matrix. Let us write Eq. (A.2) for each component of the vector  $\mathbf{x}$ :

$$x_i(t) = \lim_{m \rightarrow \infty} \sum_{i_1, \dots, i_{m-1}, j=1}^n (\delta_{ii_1} + A_{ii_1} dt) \dots (\delta_{i_{m-1}j} + A_{i_{m-1}j} dt) \bar{x}_j \quad (A.3)$$

The regrouping of terms in this equation yields

$$x_i(t) = \lim_{m \rightarrow \infty} \left[ (1 + A_{ii} dt)^m \bar{x}_i + \sum_{l=0}^m \sum_{\substack{k_0, k_1, \dots, k_l=1 \\ k_p \neq k_{p-1}, p=1, \dots, l \\ k_l \neq i}}^n \sum_{\substack{m_0, \dots, m_l \geq 1 \\ m_0 + \dots + m_l \leq m}} (1 + A_{ii} dt) \times m - \sum_{p=0}^l m_p A_{ik_l} dt \cdot (1 + A_{k_l k_l} dt)^{m_l-1} \times A_{k_l k_{l-1}} dt (1 + A_{k_{l-1} k_{l-1}} dt)^{m_{l-1}-1} \dots A_{k_1 k_0} dt (1 + A_{k_0 k_0} dt)^{m_0-1} \bar{x}_{k_0} \right] \quad (A.4)$$

Written explicitly in Eq. (A.4) are the values of the Kronecker deltas  $\delta_{qr}$  that appear in Eq. (A.3), and the resulting terms are grouped according to the number of the nondiagonal elements of matrix  $\mathbf{A}$ . Two external sums in Eq. (A.4) sort out all the ordered sequences of the indices, i.e., the "trajectories"  $k_0 k_1 \dots k_l i$ , where  $k_0$  and  $i$  are its initial and final points, while  $k_1, \dots, k_l$  are the consecutive intermediate points. The separate term corresponds to a single-point trajectory and with  $m \rightarrow \infty$  it tends to  $\bar{x}_i \exp(A_{ii}t)$ .

The internal sum in Eq. (A.4), with  $m \rightarrow \infty$ , tends to the solution of the  $(l + 2)$ 'th order set similar to



Eq. (A.1) but having a two-diagonal matrix  $\mathbf{B}$  consisting of matrix  $\mathbf{A}$  elements:

$$B_{pp} = A_{k_p k_p}, \quad p = 0, \dots, l, B_{l+1, l+1} = A_{ii}$$

and

$$B_{p, p-1} = A_{k_p k_{p-1}}, \quad p = 1, \dots, l, B_{l+1, l} = A_{ik_l} \quad (\text{A.5})$$

and the initial condition  $Y_p(0) = \tilde{Y}_p = \delta_{p0} \tilde{x}_{k_0}$ . Indeed, taking into account the relationship

$$(1 + A_{k_l k_l} dt)^{m_l-1} \xrightarrow{m \rightarrow \infty} \exp[A_{k_l k_l} (m_l - 1) dt] \quad (\text{A.6})$$

and denoting  $t_p = (m_p - 1) dt$ , one can see that with  $m \rightarrow \infty$  the internal sum in Eq. (A.4) is transformed into the expression

$$Y_i = \tilde{Y}_0 \int_{t_0 \dots t_l \geq 0} \dots \int \theta(t - t_0 - \dots - t_l) \times \prod_{p=0}^l [B_{p+1, p} \cdot dt_p \cdot \exp(B_{pp} t_p)] \times \exp[B_{l+1, l+1} (t - t_0 - \dots - t_l)] \quad (\text{A.7})$$

which is similar to Eq. (10) and yields the solution to the problem

$$\frac{dY_p}{dt} = B_{p, p-1} Y_{p-1}(t) + B_{pp} Y_p(t) \quad (\text{A.8})$$

$$Y_p(0) = \delta_{p0} \tilde{x}_{k_0} \quad (\text{A.8})$$

Taking into account Eqs. (1) and (2), one can write Eq. (A.4) in the form

$$x_i(t) = \tilde{x}_i \cdot \exp(A_{ii} t) + \sum_{l=0}^{\infty} \sum_{\substack{k_0, k_1, \dots, k_l=1 \\ k_p \neq k_{p-1}, k_l \neq i}}^n (A_{k_p k_p} t) \times \tilde{x}_{k_0} \left[ \prod_{p=0}^{l-1} (A_{k_{p+1}, k_p} t) \right] (A_{ik_l} t) \times D_{l+2}(-A_{k_0 k_0} t, \dots, -A_{k_l k_l} t, -A_{ii} t) \quad (\text{A.9})$$

If matrix  $\mathbf{A}$  is time dependent, similar transformations lead to an expression of the type in Eq. (9), which is also the solution to the problem in Eq. (A.8). When matrix  $\mathbf{A}$  is triangular, the indices  $k_0, \dots, k_l$  in Eq. (A.4) are increase ordered, i.e.,  $k_0 < k_1 < \dots < k_l$ , the order of matrix  $\mathbf{B}$  does not exceed  $n$ , and the number of considered trajectories is finite. When matrix  $\mathbf{A}$  is nontriangular, the series in  $l$  becomes infinite, but it can be easily verified that it converges absolutely for any finite value of  $t$ .

Equation (A.9) can be written as

$$x_i(t) = P_{ij}(t) x_j \quad (\text{A.10})$$

where  $P_{ij}(t)$  is the matricant, or the propagator of Eq. (A.1) (Refs. 12 and 13), and  $x_j$  are the initial conditions. Equation (A.9) provides representation of  $P_{ij}(t)$  as a sum of contributions by the trajectories  $j, k_j, \dots, k_l, i$ , each contribution being found by solv-

ing the "two-diagonal" problems of Eqs. (A.5) and (A.8). Note that unlike the splitting methods expressing the solution of Eq. (A.1) through eigenvalues of matrix  $\hat{\mathbf{A}}$  (Ref. 13), here  $P_{ij}(t)$  comprises only elements of this matrix. Such a representation of the matricant of a set of the ordinary differential equations has not, to our knowledge, been considered earlier.

## APPENDIX B

### BASIC FORMULAS FOR DEPLETION FUNCTIONS

Given in this Appendix are basic relationships for the depletion functions [Eq. (3)]. The proofs, additional formulas, and the FORTRAN-IV subroutine DEPLET that permits computation of  $D_n(x_1, \dots, x_n)$  with  $n \leq 15$  for any real values of the arguments can be found in Ref. 7.

The recurrent relationships are

$$D_1(x_i) = \exp(-x_i)$$

$$D_n(x_1, \dots, x_n) = \frac{D_{n-1}(x_1, \dots, x_{i-1}, x_{i+1}, \dots, x_n)}{x_i - x_j} - \frac{D_{n-1}(x_1, \dots, x_{j-1}, x_{j+1}, \dots, x_n)}{x_i - x_j} \quad (\text{B.1})$$

Hence, the expression for the partial derivative is

$$\frac{\partial}{\partial x_i} D_n(x_1, \dots, x_n) = -D_{n+1}(x_1, \dots, x_n, x_i) \quad (\text{B.2})$$

The integral representation is

$$D_n(x_1, \dots, x_n) = \int_{t_1, \dots, t_n \geq 0} \dots \int dt_1 \dots dt_n \delta(1 - t_1 - \dots - t_n) \times \exp[-(t_1 x_1 + \dots + t_n x_n)] \quad (\text{B.3})$$

which makes it clear that the depletion functions are symmetrical with respect to all their arguments. Unlike Eq. (3), the representation of Eq. (B.3) is also applicable when the values of some arguments are identical. The following relationships follow from Eqs. (B.1) and (B.3):

$$D_n(\{x_1\}_n) = \frac{\exp(-x_1)}{(n-1)!} \quad (\text{B.4})$$

and

$$D_n(\overbrace{x_1, \dots, x_1}^{n-1}, x_2) = \exp(-x_2) \gamma^*(n-1, x_1 - x_2) \quad (\text{B.5})$$

Here,  $\gamma^*(m, x)$  is the modified incomplete gamma function  $\gamma^*(m, x) = x^{-m} \gamma(m, x) / (m - 1)!$  (Ref. 21) (see also Refs. 7 and 8).

If all the arguments  $x_1, \dots, x_n$  have closely spaced values, the depletion function can be expanded into a fast-converging power series of  $x_i - x_0$  where  $x_0 = (x_1 + \dots + x_n) / n$

$$D_n(x_1, \dots, x_n) = \frac{\exp(-x_0)}{(n-1)!} \left[ 1 + \frac{s_2}{2n(n+1)} - \frac{s_3}{3n(n+1)(n+2)} + \frac{2s_4 + s_2}{8n \dots (n+3)} - \frac{6s_5 + 5s_2s_3}{30n \dots (n+4)} + \dots \right], \quad (B.6)$$

where  $s_k = (x_1 - x_0)^k + \dots + (x_n - x_0)^k$ .

For any real values of the arguments, the following inequalities hold true:

$$\frac{\exp(-x_0)}{(n-1)!} \leq D_n(x_1, \dots, x_n) \leq \frac{1}{n!} \sum_{i=1}^n \exp(-x_i). \quad (B.7)$$

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