ANALYTICAL METHOD FOR SOLVING RADIOACTIVE TRANSFORMATIONS

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1. INTRODUCTION

Nuclide concentrations N_i, $i=1, \ldots, n$, in a single linear radioactive chain satisfies the equations:

$$\frac{dN}{dt} = s_{i-1}N_{i-1} - \lambda_i N_i \tag{1}$$

where:

l_i - total depletion constant of the i-th nuclide;

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

Under initial conditions

$N_1(t=0)=N_{10}$, $N_i(t=0)=0$; $i=2, \ldots, n$,

well-known Bateman soluton [1] of Eq. (1) can be written in the form (2):

$$N_n(t) = N_{10}\sum_{i=1}^{n} B_n^i \cdot \exp(\lambda_i t) \tag{2}$$

where

$$B_n^i = \frac{s_i}{s_n}\prod_{\substack{j=1\\ j\neq i}}^{n}\frac{s_j}{\lambda_j - \lambda_i} \tag{2a}$$

In spite of its simplicity Eq. (2) is not always convenient for the calculation of the nuclide concentrations. When the differences $(\lambda_j-\lambda_i)$ in the denominator of the Bateman coefficients, Eq. (2a), become small, the round-off error in evaluating the Bateman sum, the right-hand sum in Eq. (2), numerically in a finite-precision calculation invalidates further computation. However, if some of the arguments l_i are equal, the Bateman coefficients can not be computed.

2. DESCRIPTION OF THE METHOD

The depletion function method was developed to overcome numerical difficulties [2-8]. Defining depletion function of k arguments as (3):

$$F_k(x_k,\ldots,x_1) = \sum_{j=1}^{k}\exp(-x_j)\prod_{\substack{i=1\\ i\neq j}}^{k}\frac{1}{x_i - x_j} \tag{3}$$

the solution of Eq. (1) become (4):

$$N_n(t) = N_{10}\left[\prod_{i=1}^{n-1} s_i t\right]F_n(x_n,\ldots,x_1) \tag{4}$$

where $x_i=l_i t$; $i=1, \ldots, n$.

The function F_k is symmetric in all its arguments and the recurrence relation holds (5):

$$F_k(x_k,\ldots x_1) = \frac{F_{k-1}(x_k,\ldots x_3,x_1) - F_{k-1}(x_k,\ldots,x_3,x_2)}{x_2 - x_1} \tag{5}$$

The expression for the power series of the depletion function must be used in the case where all the arguments x_i have closely spaced or equal values [7]:

$$F_k(x_k,\ldots,x_1) = F_1(x_k)A_k \tag{6}$$

$$A_k = \sum_{i=k-1}^{n}\frac{1}{i!}S_i^{(k)} \tag{7}$$

$$S_i^{(k)} = \sum_{m=k-2}^{i-1}\binom{i-1}{m}X_{k,k-1}^{i-1-m}\,S_m^{(k-1)} \tag{8}$$

where:

$x_{k,k-1} = x_k - x_{k-1}, \ldots, x_{2,1} = x_2 - x_1$

and

$S_1^{(2)} = 1; S_2^{(2)} = x_{2,1}; \ldots; S_2^{(n)} = x_{2,1}^{n-1}$

The expression for the relative error (9) [8]:

$$\delta_r \leq \frac{(k-1)(k-1)!\,\varepsilon^{\,n-k+2}}{[n+2-(\varepsilon+1)(k-1)]n!}\,\Im_n^{k-1} \tag{9}$$

where $\Im_n^{k-1}$ is the Stirling number of the second kind, has to be used to calculate the depletion function power series to a desired accuracy if the series (7) is cut at the n-th term and if $x_{i,i-1}\leq\varepsilon$; $i=2, \ldots, k$. It is proved [8] that that the power series of the depletion function is fast converging one for the small values of $\varepsilon(\varepsilon\leq 0.1)$, even for the case that $\delta_r\leq 10^{-16}$.

If some of the differences $x_{k,k-1}= x_k - x_{k-1}$ are too small, one can calculate the depletion function to good accuracy using recurrence relation (5). But, if the number of nuclides in the chain is too large, the number of operations become enormous, and the calculation is practically impossible. To calculate the concentrations of the nuclide the method of nonsingular Bateman coefficients (NBCs) is developed [9].

Let, of the n arguments of the function $F_n(x_1, \ldots, x_n)$, $n(1)$ arguments have values closely spaced to the value $x(1)$ (1st group), $n(2)$ arguments have values closely spaced to the value $x(2)$ (2nd group),... , $n(k)$ arguments have values closely spaced to the value $x(k)$ (k-th group); with $n(1)+n(2)+\ldots+n(k)=n$, where the values $x(1)$, $x(2)$, ..., $x(k)$ differ significantly between each other. Let $m(k)$ be:

$$m(k) = \sum_{i=1}^{k} n(i)\,,\ i=1, \ldots, k;\ n(0)=0.$$

It is proved [9] that the depletion function F_n can be evaluated using expression (10):

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$$F_n(x_1,\ldots,x_n) = \sum_{k=1}^{K}\sum_{i=1}^{n(k)} F_i[x_{m(k-1)+1},\ldots,x_{m(k-1)-i}) * K_{n(k),n}^{n(k)-i+1}[x_{m(k-1)+i},\ldots,x_{m(k)}] \quad (10)$$

with the definition of nonsingular Bateman coefficients:

$$K_{M,M}^{1} = 1;$$

$$K_{M,M}^{i} = 0,\ i = 2,\ldots M;$$

$$K_{M,M+s}^{0} = 0,\ s = 1,\ldots,n-M; \quad (11)$$

$$K_{M,M+s}^{i}(x_{M-i+1},x_{M-i+2},\ldots,x_M) = \frac{1}{x_{M+s}-x_{M-i+1}}$$

$$*[K_{M.M+s-1}^{i}(x_{M-i+1},\ldots,x_M) - K_{M,M+s}^{i-1}(x_{M-i+2},\ldots,x_M)]$$

By successive application of Eq. (11) one can obtain all the NBCs $K_{M,n}^{i}$, i=1, ..., M. Differences of the arguments of the same group can never be used. Therefore, the NBCs can have no singularities.

When all the arguments of the depletion function F_n are significantly different NBCs reduces to Bateman coefficients and the Eq. (4) comes to a Bateman solution. When all the arguments of the depletion function F_n are equal or near equal, F_n is calculated using its power series expansion.

3. EXAMPLES

Numerous calculations have been performed on the simple radioactive transformations with up to 100 consecutive nuclides. The last nuclide was stable. In such chains, the sum of all nuclide concentrations in the chain is equal to the initial nuclide concentrations, and this fact is used to check the accuracy of the calculation. The decay constants were artificially chosen to obtain a different number of groups with equal or near equal arguments. In a number of cases where the Bateman solution and the depletion functions solution failed, the exact solution was obtained to good or even high accuracy. One of these examples is presented here to demonstrate the ability of the method. Figure 1 shows the concentration of the nuclide 30 in a dummy chain where:

- nuclides 1, 2, 6, 7, 13, 18, 26, and 30 have decay constants from 7.0 s^{-1} to 7.3 s^{-1},
- nuclides 8, 9, 12, 20, 24, and 28 have decay constants from 17.2 s^{-1} to 17.4 s^{-1},
- nuclides 22, 23, and 29 have decay constants from 20.0 s^{-1} to 20.2 s^{-1},
- nuclides 10, 11, 15, and 21 have decay constants from 25.0 s^{-1} to 25.3 s^{-1},
- nuclides 3, 4, 5, 17, and 27 have decay constants from 35.0 s^{-1} to 35.3 s^{-1},
- nuclides 14, 16, and 29 have decay constants from 117.0 s^{-1} to 117.1 s^{-1},
- nuclide 25 has decay constant of 300.0 s^{-1}.

Concentration of the nuclide 30 has been calculated using Bateman solution and the NBC method and it is a good illustration of a round-off errors that occur when applying Bateman solution. It, also, demonstrates the high accuracy of the NBC method.

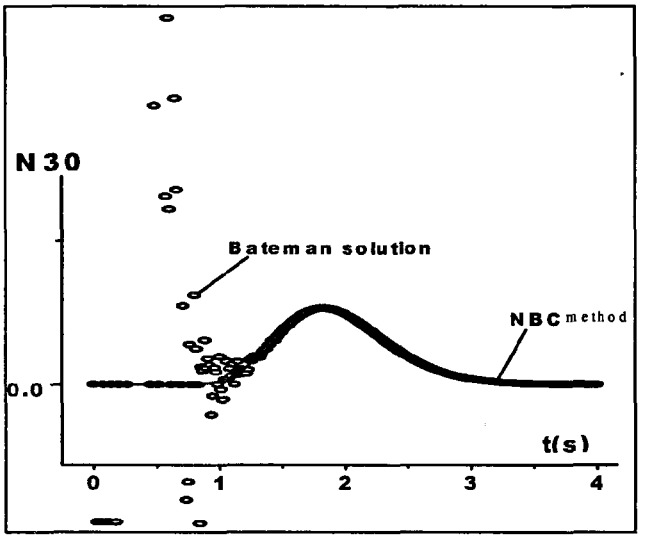

Figure 1. Concentration of the 30th nuclide in a dummy nuclide chain. o - Bateman solution; ▬ - NBC method

4. SUMMMARY

The exact method of solving radioactive transformations is presented. Nonsingular Bateman coefficients, which can be computed using recurrence formulas, greatly reduce computational time and eliminate singularities that often arise in problems involving nuclide transmutations. Depletion function power series expansion enables high accuracy of the performed calculations, specially in a case of a decay constants with closely spaced values. Generality and simplicity of the method make the method useful for many practical applications.

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