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Pulsed/Intermittent Activation within Fusion  
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Within Fusion Energy Devices**

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## **Abstract**

Two calculational methods have been developed to compute the induced radioactivity due to pulsed/intermittent irradiation histories as encountered in both magnetic and inertial fusion energy devices. The numerical algorithms are based on the linear chain method (Bateman Equations) and employ series reduction and matrix algebra. The first method models the case in which the irradiated materials are present throughout the series of irradiation pulses. The second method treats the case where a fixed amount of radioactive and transmuted material is created during each pulse. Analytical solutions are given for each method for a three nuclide linear chain. Numerical results and comparisons are presented for a select number of linear chains.

# I Introduction

The nuclear analysis of experimental and commercial fusion devices requires the calculation of radioactivity and related parameters such as afterheat, dose rates, waste disposal ratings, and biological hazard potential. These quantities play an important role in determining the first wall material, blanket structure, environmental impact, economics, maintenance procedures, and radiation exposure to personnel from harmful radiation of fusion devices. The major computational tool employed for the radioactivity calculations is a transmutation/activation code. This code computes the nuclear transmutation of the initial stable nuclides and the nuclear and radioactive decay transmutations of the subsequent secondary stable and radioactive nuclides. It is this computational tool and its application to the problem of pulsed/intermittent activation, as encountered in fusion energy devices, which is the subject of this discussion.

Application of pulsed or intermittent irradiation operation within fusion energy systems has previously been encountered in several experimental/test facilities and reactor studies. The calculation of biological dose rates for the Tokamak Fusion Test Reactor [1] was examined for three pulse schedules: (1) 100 consecutive pulses in a day, (2) 100 consecutive pulses each day for 10 consecutive days, and (3) 100 consecutive pulses each day operating with a 10 day shutdown period between operating days. Another study examined biological dose rates at the ion diode of the proposed Light Ion Fusion Target Development Facility [2]. This study utilized the pulse schedule depicted in Figure 1 which consists of 12 shots per day, 5 days a week, 52 weeks a year. A similar pulsing schedule has been suggested for the Laboratory Microfusion Facility [3]. A total of 500 shots a year are considered and are distributed over a 50 week time period. The operation schedule allows for 2 shots per day which are 6 hours apart with 18 hours between the last shot on the first day and the first shot on the second day. Operating for 5 days a week results in 10 shots a week. The weekly pulse sequences are 66 hours apart. Another example is the activation analysis performed for the replacement of HT-9 steel by low activation modified HT-9 steel for the LIBRA fusion reactor study [4]. The pulse sequence used in this study is shown in Figure 2. The reactor is assumed to be shut down for 5 days every month for routine maintenance and for 40 days every year for extended maintenance. The tentative operation mode of the International Thermonuclear Experimental Reactor (ITER) is projected to be intermittent with pulse widths ranging from several minutes to an hour and off times of several minutes [5].

In the cases cited above, the calculation of the biological dose rates was performed using various numerical models which approximate the pulsed/intermittent irradiation history of the fusion devices. The approximate methods fall into two categories: those methods which use a steady state (continuous irradiation) treatment of the irradiation history and those methods which preserve the pulsed nature of the irradiation history. A

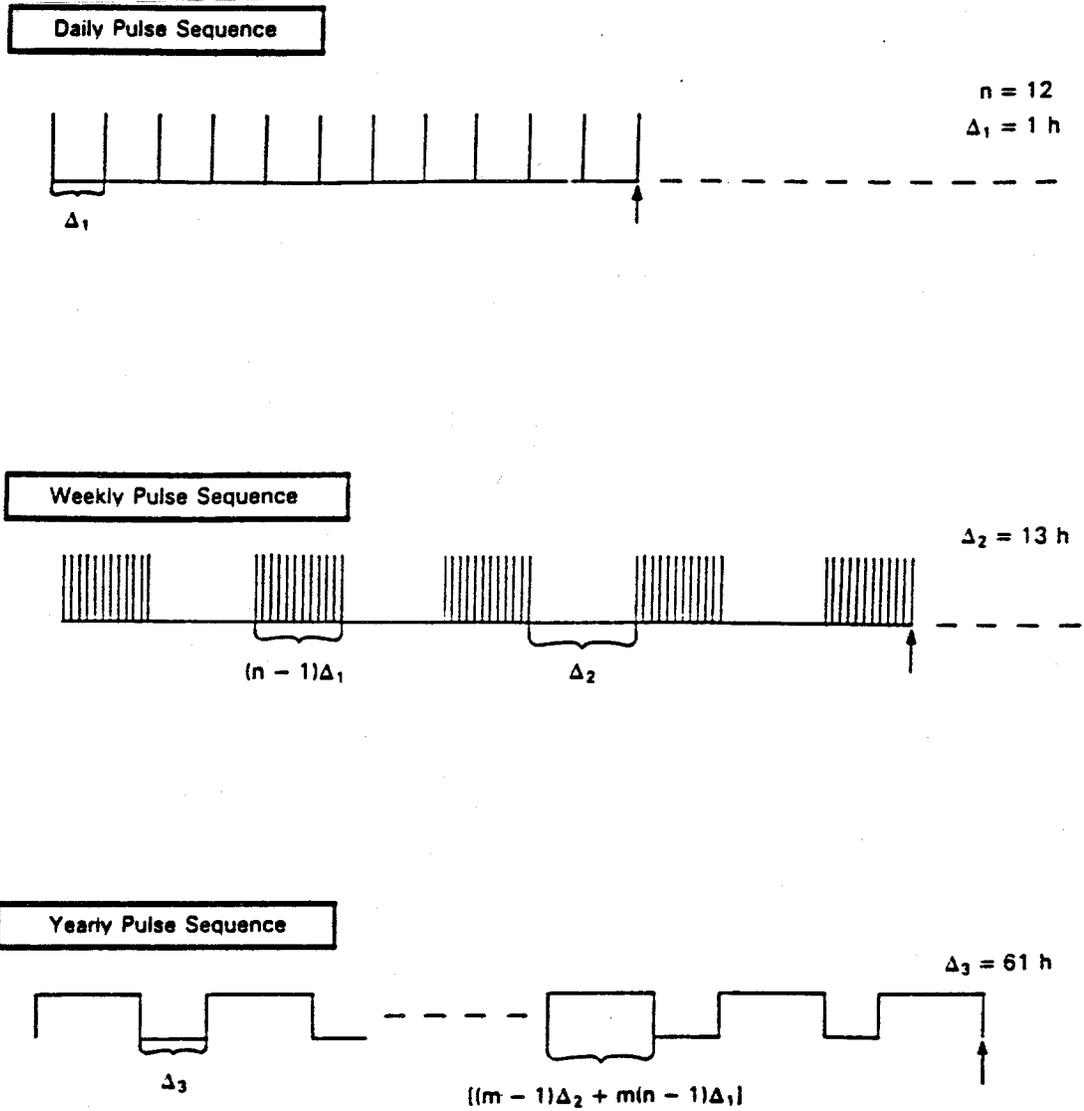


Figure 1. Pulse sequence schedule used for the proposed Light Ion Fusion Target Development Facility.

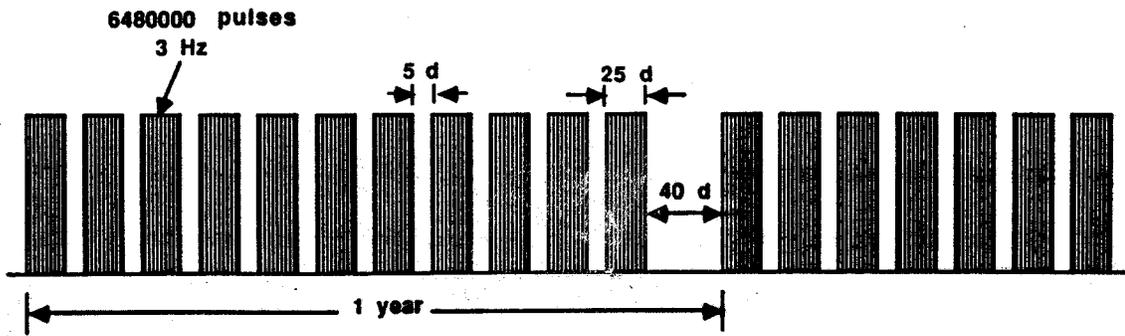


Figure 2. Pulse sequence schedule used in the LIBRA reactor study.

detailed review of the steady state methods employed in the activation analysis of inertial fusion energy (IFE) and magnetic fusion energy (MFE) reactors can be found in ref. 6. Depending on the averaging technique used for the flux intensity, pulse width and off time, it was shown that the steady state methods can over-/under- estimate the activity and hence the biological dose rate by orders of magnitude. For the more realistic methods which preserve the pulsed nature of irradiation history, the basic assumption used is that the destruction of the initial and transmuted/activated nuclides by neutron interactions in the second and subsequent pulses is the same as that during the first pulse [7,8,9]. That is, the approximation assumes the creation and destruction of the same amount of radioactive and stable nuclides during each pulse. Activation/transmutation models based on this approximation have been employed in several experimental and power reactor studies [1-5,7] and produce good results provided the destruction rate is small, and the initial stable nuclide is transmuted to a radioactive one. Significant errors may occur for long operating times and for nuclides beyond the second in a transmutation chain [10]. Method B developed in this paper is an improvement to these realistic methods. A comparison with the exact method for the calculation of the pulse history (Method A developed in this paper) illustrates the limitations of these methods.

We note that theoretically any steady state code should be able to perform pulsed/intermittent activation calculations without having to resort to approximate methods. One simply repeatedly executes the steady state code for each pulse width and decay period using the calculational results of the previous period as initial conditions for the current period. This process can be automated; however it can be quite time consuming, especially if there is interest in computing several thousands to tens of thousands of pulses for numerous nuclides as is the case for fusion energy systems. Hence the above procedure is quite inefficient for large numbers of pulses. The fission reactor isotope depletion code ORIGEN2 [11] and the THIDA-2 transmutation, decay heat and

dose rate code system [12] can treat pulse/intermittent histories for a small number of pulses. However the numerical routines employed in these codes are computationally inefficient for the large number of pulses encountered in fusion systems.

Two computationally efficient methods have been developed to compute the induced activity due to pulsed/intermittent irradiation histories in fusion energy devices. The first method treats the case in which the irradiated materials are present throughout the series of irradiation pulses [10]. This method explicitly accounts for the further destruction of the initial and transmuted materials during the second and subsequent pulses, contains no approximations, and applies to any structural material (first wall, magnet, shield, etc.) which is present in the reactor for its lifetime. The material is exposed to the total reactor irradiation history including pulses and maintenance periods. The second method models the case where the initial material concentrations are present at the beginning of each pulse which results in the same amount of radioactive and transmuted material generated during each pulse. This method is exact for the case where the transmuted and activated materials are extracted after each pulse to avoid further irradiation of the activated materials.

In Section II, we briefly review the basic transmutation rate equations which govern the time rate of change of the stable and radioactive nuclide densities. The solution of the rate equations by the linear chain method is introduced and is formulated in terms of a lower triangular matrix. Use is made of the lower triangular matrix formulation to account for burnup, production, and decay of nuclides during pulsed/intermittent irradiation in the development of the computational methods. Solution algorithms are developed using series reduction and matrix algebra. Analytical solutions for a three nuclide linear chain are presented for each computational method. Results for a select number of linear chains are presented in Section III. This is followed by a brief summary given in Section IV.

## **II Mathematical Formulation - Theory**

### **II.A The Transmutation Rate Equations**

The transmutation rate equations are a system of coupled first order linear differential equations which govern the time rate of change of nuclide densities resulting from the transmutation/activation of nuclides by neutron irradiation. The equations are of the form

$$\begin{aligned} \frac{dN_i(\vec{r}, t)}{dt} = & \Phi(\vec{r}, t) \times \sum_j N_j(\vec{r}, t) \sigma_{j \rightarrow i}(\vec{r}) + \sum_k N_k(\vec{r}, t) \lambda_{k \rightarrow i} \\ & - \Phi(\vec{r}, t) N_i(\vec{r}, t) \times \sum_\ell \sigma_{i \rightarrow \ell}(\vec{r}) - N_i(\vec{r}, t) \sum_m \lambda_{i \rightarrow m} + S_i(\vec{r}, t). \end{aligned} \quad (1)$$

where:

$$\begin{aligned} N_i(\vec{r}, t) &= \text{number density of nuclide } i \text{ at time } t, \text{ at position } \vec{r}; \\ \Phi(\vec{r}, t) &= \text{total flux at time } t, \text{ at position } \vec{r}; \\ \sigma_{n \rightarrow p}(\vec{r}) &= \text{microscopic cross section for changing nuclide } n \text{ into nuclide } p \text{ at position } \vec{r}; \\ \lambda_{q \rightarrow s} &= \text{decay constant for nuclide } q \text{ changing into nuclide } s; \\ S_i(\vec{r}, t) &= \text{external source of nuclide } i \text{ at time } t, \text{ at position } \vec{r}; \end{aligned}$$

and where all references to multigroup fluxes and spectra averaged cross sections have been suppressed. The initial conditions are that  $N_i(\vec{r}, 0) = N_{i0}(\vec{r})$ .

Equation 1 can be cast into a general matrix form (where the spatial variable has been suppressed);

$$\frac{d\bar{N}}{dt} = \mathbf{A} \bar{N}(t) + \bar{S}(t) \quad (2)$$

where:

$$\begin{aligned} \bar{N}(t) &= \text{nuclide density vector} = [N_1(t), N_2(t), \dots, N_N(t)]^T; \\ \bar{S}(t) &= \text{external source vector} = [S_1(t), S_2(t), \dots, S_N(t)]^T; \\ \mathbf{A} &= \text{matrix} = \{a_{ij}\}; \\ a_{ij} &= -(\sigma_i^D \Phi + \lambda_i) \delta_{ij} + (\sigma_{j \rightarrow i}^F \Phi + \lambda_j b_{j \rightarrow i}); \\ \sigma_i^D &= \text{microscopic destruction cross section of nuclide } i; \\ \sigma_{j \rightarrow i}^F &= \text{microscopic production cross section of nuclide } j \text{ forming nuclide } i; \\ \lambda_i &= \text{decay constant of nuclide } i; \\ b_{j \rightarrow i} &= \text{branching ratio of a decay from nuclide } j \text{ to nuclide } i. \\ \delta_{ij} &= \text{the Kronecker delta.} \end{aligned}$$

Several numerical algorithms have been developed for the solution of large matrix systems as obtained in the general matrix form. The methods utilized are called the Matrix Exponential Method [13,14], the Gear method [15-17], the Adams method [18], the Runge-Kutta methods [19], the Fine Step Forward Differencing method [20], the Eigenvalue method [21], the Avalanche method [22] and most recently the Perturbation method [23].

Alternatively, the transmutation rate equations can be cast into a coupled set of linearized decay/transmutation chain equations where the number density of the  $i$ 'th nuclide is related to the preceding nuclide in the chain;

$$\frac{dN_i(t)}{dt} = p_{i-1 \rightarrow i} N_{i-1}(t) - d_{i \rightarrow i+1} N_i(t) + S_i(t) \quad (3)$$

where:

$$\begin{aligned} p_{i-1 \rightarrow i} &= \text{the partial production rate of the } i\text{'th nuclide from the } i\text{'th-1 nuclide;} \\ d_{i \rightarrow i+1} &= \text{the destruction rate of the } i\text{'th nuclide.} \end{aligned}$$

The decomposition of the general matrix form into a coupled set of linearized chains is accomplished through the separation of the various production modes of a nuclide into a set of equations describing the partial nuclide concentration due to a single decay or neutron transmutation mode. Thus each equation contains only a single production mode but all modes of loss (destruction). The number of linearized chains generated for the partial concentration of the  $i$ 'th nuclide is equal to the number of production modes which lead to the  $i$ 'th nuclide. Each linearized chain is independent and hence the partial concentrations can be computed. The concentrations for a given nuclide, whether radioactive or stable, can be obtained by adding the contributions to the nuclide from the various linearized chains which contain the nuclide of interest.

The system of linearized equations have an analytical solution known as the Bateman Equations [24,25]. We note that these equations are exact and do not rely on numerical approximations. The solution for the  $i$ 'th member of a chain at a time  $t$  can be written in the form [11]:

$$\begin{aligned} N_i(t) &= N_i(0) e^{-d_i t} \\ &+ \sum_{k=1}^{i-1} N_k(0) \left[ \sum_{j=k}^{i-1} \frac{P_{j+1,j} (e^{-d_j t} - e^{-d_i t})}{(d_i - d_j)} \prod_{n=k, n \neq j}^{i-1} \frac{P_{n+1,n}}{d_n - d_j} \right] \end{aligned}$$

where:

$$\begin{aligned} P_{i+1,i} &= \text{production rate of the } i\text{'th+1st member from the } i\text{'th member} = \sigma\Phi \text{ or } \lambda; \\ d_i &= \text{destruction rate of the } i\text{'th member} = (\sigma\Phi + \lambda), \sigma\Phi, \text{ or } \lambda; \\ N_i(0) &= \text{initial number density of nuclide } i. \end{aligned}$$

For a three nuclide chain the above general form reduces to

$$\begin{aligned}
N_1(t) &= N_1(0) e^{-d_1 t} \\
N_2(t) &= N_2(0) e^{-d_2 t} + N_1(0) \frac{P_2(e^{-d_1 t} - e^{-d_2 t})}{(d_2 - d_1)} \\
N_3(t) &= N_3(0) e^{-d_3 t} + N_2(0) \frac{P_3(e^{-d_2 t} - e^{-d_3 t})}{(d_3 - d_2)} \\
&\quad + N_1(0) \frac{P_2 P_3}{(d_2 - d_1)} \frac{(e^{-d_1 t} - e^{-d_3 t})}{(d_3 - d_1)} + N_1(0) \frac{P_2 P_3}{(d_1 - d_2)} \frac{(e^{-d_2 t} - e^{-d_3 t})}{(d_3 - d_2)},
\end{aligned}$$

where:

$$\begin{aligned}
P_2 &= P_{2,1} ; \\
P_3 &= P_{3,2} .
\end{aligned}$$

The solution to the linearized chain method can be written in lower triangular matrix form. The general structure of the lower triangular matrix can be gleaned from the solution of the above three nuclide chain;

$$\begin{bmatrix} N_1(t) \\ N_2(t) \\ N_3(t) \end{bmatrix} = \begin{bmatrix} e^{-d_1 t} & 0.0 & 0.0 \\ \frac{P_2 (e^{-d_1 t} - e^{-d_2 t})}{(d_2 - d_1)} & e^{-d_2 t} & 0.0 \\ \frac{P_2 P_3}{(d_2 - d_1)} \frac{(e^{-d_1 t} - e^{-d_3 t})}{(d_3 - d_1)} + \frac{P_3 (e^{-d_2 t} - e^{-d_3 t})}{(d_3 - d_2)} & & e^{-d_3 t} \\ + \frac{P_2 P_3}{(d_1 - d_2)} \frac{(e^{-d_2 t} - e^{-d_3 t})}{(d_3 - d_2)} & & \end{bmatrix} \begin{bmatrix} N_1(0) \\ N_2(0) \\ N_3(0) \end{bmatrix} .$$

Writing the above in general matrix form we have

$$\bar{N}(t) = \mathbf{B} \bar{N}(0) . \tag{4}$$

The matrix form of the linear chain solution is the central (key) component in the following development of the pulsed/intermittent computational methods.

## II.B Pulsed/Intermittent Activation

For simplicity in the development of the calculational schemes, it is assumed that we have a series of uniform pulses having a pulse width of  $\Delta t_1$  and a dwell time (off time) of  $\Delta t_2$  and that the irradiation flux level is the same for all pulses. The pulse

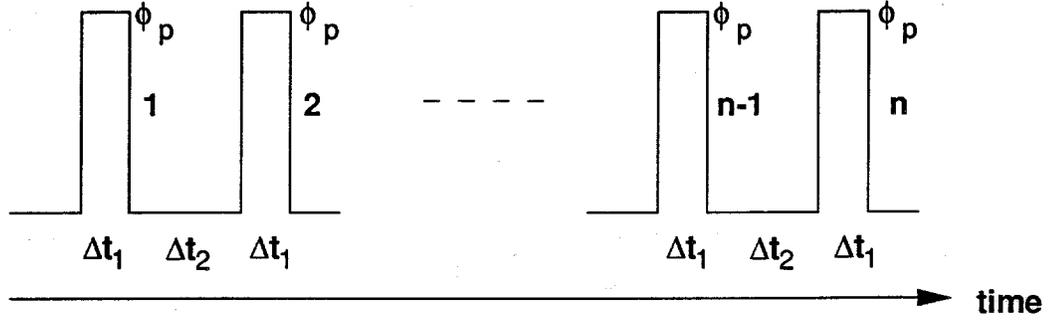


Figure 3. A series of uniformly spaced pulses of width  $\Delta t_1$ , dwell (off) time  $\Delta t_2$  and flux level  $\phi_p$ .

history is depicted in Fig. 3. Computational methods based on irradiation histories with varying flux levels and varying pulsing schedules and maintenance periods have also been developed [6]. Illustrations of nonuniform pulsing schedules which can be modeled are depicted in Figs. 1, 2 and 4.

Examining the pulsed problem, we note that there are actually two processes occurring, irradiation (destruction) and decay. Each process has its individual solution which can be written in lower triangular matrix form;

$$\text{DESTR} = \begin{bmatrix} e^{-d_1 \Delta t_1} & 0.0 & 0.0 \\ \frac{P_2 (e^{-d_1 \Delta t_1} - e^{-d_2 \Delta t_1})}{(d_2 - d_1)} & e^{-d_2 \Delta t_1} & 0.0 \\ \frac{P_2 P_3}{(d_2 - d_1)} \frac{(e^{-d_1 \Delta t_1} - e^{-d_3 \Delta t_1})}{(d_3 - d_1)} + \frac{P_3 (e^{-d_2 \Delta t_1} - e^{-d_3 \Delta t_1})}{(d_3 - d_2)} & & e^{-d_3 \Delta t_1} \\ + \frac{P_2 P_3}{(d_1 - d_2)} \frac{(e^{-d_2 \Delta t_1} - e^{-d_3 \Delta t_1})}{(d_3 - d_2)} & & \end{bmatrix}$$

and

$$\text{DECAY} = \begin{bmatrix} e^{-\lambda_1 \Delta t_2} & 0.0 & 0.0 \\ \frac{\lambda_1 (e^{-\lambda_1 \Delta t_2} - e^{-\lambda_2 \Delta t_2})}{(\lambda_2 - \lambda_1)} & e^{-\lambda_2 \Delta t_2} & 0.0 \\ \frac{\lambda_1 \lambda_2}{(\lambda_2 - \lambda_1)} \frac{(e^{-\lambda_1 \Delta t_2} - e^{-\lambda_3 \Delta t_2})}{(\lambda_3 - \lambda_1)} + \frac{\lambda_2 (e^{-\lambda_2 \Delta t_2} - e^{-\lambda_3 \Delta t_2})}{(\lambda_3 - \lambda_2)} & & e^{-\lambda_3 \Delta t_2} \\ + \frac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)} \frac{(e^{-\lambda_2 \Delta t_2} - e^{-\lambda_3 \Delta t_2})}{(\lambda_3 - \lambda_2)} & & \end{bmatrix}$$

where:

- DESTR** = the destruction matrix representing nuclide destruction during the pulse;
- DECAY** = the nuclide decay matrix representing nuclide decay during the dwell (off) time;
- and  $\lambda_i$  = the decay constant for nuclide  $i$ .

The size of these matrices is dependent on the number of nuclides considered in the chain. Use of the above matrices will now be made in the development of the computational methods for activation calculations due to pulsed/intermittent irradiation.

### II.B1. Method A: Continuous Destruction/Production of Nuclide

For the case where the irradiated materials are present throughout the series of irradiation pulses, we find that after one pulse the solution can be written as

$$\bar{N}(\Delta t_1) = \mathbf{DESTR}(\Delta t_1) \times \bar{N}(0). \quad (5)$$

Next, after the first decay period our solution appears as

$$\begin{aligned} \bar{N}(\Delta t_1 + \Delta t_2) &= \mathbf{DECAY}(\Delta t_2) \times \bar{N}(\Delta t_1) \\ &= \mathbf{DECAY}(\Delta t_2) \times \mathbf{DESTR}(\Delta t_1) \times \bar{N}(0). \end{aligned} \quad (6)$$

After two pulse and decay periods the solution is written as

$$\begin{aligned} \bar{N}(2(\Delta t_1 + \Delta t_2)) &= \mathbf{DECAY}(\Delta t_2) \times \mathbf{DESTR}(\Delta t_1) \times \mathbf{DECAY}(\Delta t_2) \times \\ &\quad \times \mathbf{DESTR}(\Delta t_1) \times \bar{N}(0) \end{aligned} \quad (7)$$

$$= [\mathbf{DECAY}(\Delta t_2) \times \mathbf{DESTR}(\Delta t_1)]^2 \times \bar{N}(0). \quad (8)$$

By induction, it can be inferred that after  $n - 1$  pulse and decay periods the solution can be written as (supressing the  $\Delta t_1$  and  $\Delta t_2$  arguments)

$$\bar{N}((n - 1)(\Delta t_1 + \Delta t_2)) = [\mathbf{DECAY} \times \mathbf{DESTR}]^{n-1} \times \bar{N}(0). \quad (9)$$

After the  $n$ 'th pulse, Eq. 9 becomes

$$\overline{N}((n\Delta t_1 + (n-1)\Delta t_2)) = \mathbf{DESTR} \times [\mathbf{DECAY} \times \mathbf{DESTR}]^{n-1} \times \overline{N}(0). \quad (10)$$

This result presents an accurate description of the pulsed activation process as it explicitly accounts for the actual burnup of nuclei during irradiation and decay of radioactive nuclei between irradiation periods.

To apply this method, one needs to generate the **DESTR** and **DECAY** matrices and perform numerous matrix multiplications. However, using matrix multiplication to raise a matrix to a large power can become quite computationally intensive and time consuming and is quite inefficient. An alternative method is outlined in the text Applied Linear Algebra [26] and is presented below.

Recall that a matrix with a complete set of eigenvectors can be decomposed as follows;

$$\mathbf{A} = \mathbf{P}\Delta\mathbf{P}^{-1} \quad (11)$$

where

$$\begin{aligned} \mathbf{A} &= n \times n \text{ matrix;} \\ \Delta &= \text{diagonal eigenvalue matrix;} \\ \mathbf{P} &= \text{eigenvector matrix;} \\ \mathbf{P}^{-1} &= \text{inverse of } \mathbf{P}. \end{aligned}$$

Now, to calculate  $\mathbf{A}^k$  we have

$$\mathbf{A}^k = (\mathbf{P}\Delta\mathbf{P}^{-1})(\mathbf{P}\Delta\mathbf{P}^{-1})(\mathbf{P}\Delta\mathbf{P}^{-1}) \dots \quad (12)$$

repeated  $k$  times. The associative law allows the removal of the parenthesis. Since  $\mathbf{P}^{-1}\mathbf{P} = \mathbf{I}$  where  $\mathbf{I}$  is the identity matrix, the product reduces to

$$\mathbf{A}^k = \mathbf{P}\Delta^k\mathbf{P}^{-1}. \quad (13)$$

Thus, we note that to raise a matrix to the  $k$ 'th power, all that one requires is to find the eigenvalues and eigenvectors, calculate the inverse of the eigenvector matrix, raise the eigenvalue matrix to the  $k$ 'th power (a trivial operation), and multiply everything together. Applying this result to Eq. 10 yields

$$\overline{N}((n-1)(\Delta t_1 + \Delta t_2) + \Delta t_1) = \mathbf{DESTR} \times (\mathbf{P}\Delta^{n-1}\mathbf{P}^{-1}) \times \overline{N}(0) \quad (14)$$

where  $(\mathbf{P}\Delta^{n-1}\mathbf{P}^{-1}) = (\mathbf{DECAY} \times \mathbf{DESTR})^{n-1}$ .

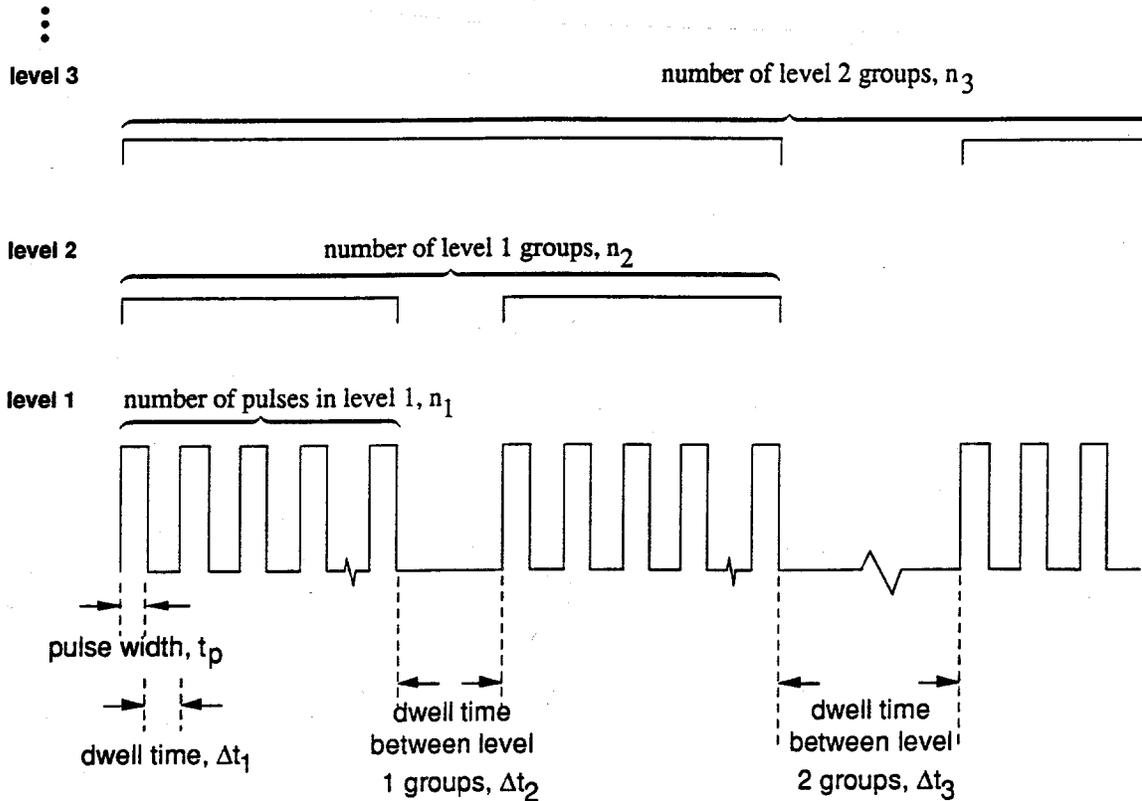


Figure 4. Relation between various levels of regular pulse groupings during pulse/intermittent irradiation.

An analytical expression for the first three nuclides within a linear chain using the method outlined above is presented in Appendix A. The above outlined method has been used to generate more complicated irradiation schedules having different pulse widths and flux levels (see Fig. 4) [6].

We note here that the construction of the **DECAY** and **DESTR** matrices is not restricted to the use of the linear chain method. Any of the other large matrix system solution techniques discussed at the beginning of Section II can be utilized. However the evaluation of Eq. 14 requires the determination of the eigenvalues and eigenvectors of the **DECAY-DESTR** product matrix and the inverse of its eigenvector matrix. The computational work involved in the determination of these matrices is greatly simplified if the **DECAY-DESTR** product matrix is lower triangular as is the case when the linear chain method (Bateman Equation solution) is utilized.

## II.B2. Method B: Constant Destruction/Production of Nuclide

For the case where the initial material concentrations are present at the beginning of each pulse resulting in the creation and destruction of the same amount of radioactive and stable nuclides during each pulse, the solution after  $n$  pulses and  $n - 1$  decay periods can be written as

$$\begin{aligned}
& \overline{N}((n\Delta t_1 + (n - 1)\Delta t_2)) \\
&= [\mathbf{DESTR}(\Delta t_1) + \mathbf{DECAY}(\Delta t_2 + \Delta t_1) \times \mathbf{DESTR}(\Delta t_1) + \\
&+ \mathbf{DECAY}(2(\Delta t_2 + \Delta t_1)) \times \mathbf{DESTR}(\Delta t_1) + \dots + \\
&+ \mathbf{DECAY}((n - 1)(\Delta t_1 + \Delta t_2)) \times \mathbf{DESTR}(\Delta t_1)] \times \overline{N}(0) \tag{15}
\end{aligned}$$

where

$$\begin{aligned}
\mathbf{DESTR} &= m \times m \text{ destruction matrix;} \\
\mathbf{DECAY} &= m \times m \text{ decay matrix;} \\
\overline{N}(0) &= \text{initial density column vector of length } m; \\
\Delta t_1 &= \text{time of one irradiation pulse;} \\
\Delta t_2 &= \text{time of one decay period;} \\
\overline{N}(n\Delta t_1 + (n - 1)\Delta t_2) &= \text{solution density column vector of length } m \text{ after } n \\
&\quad \text{pulses and } n - 1 \text{ decay periods.}
\end{aligned}$$

Since the destruction matrix,  $\mathbf{DESTR}$ , is identical for each term of the series, it can be extracted from the brackets yielding

$$\begin{aligned}
& \overline{N}((n\Delta t_1 + (n - 1)\Delta t_2)) \\
&= [\mathbf{I} + \mathbf{DECAY}(\Delta t_2 + \Delta t_1) + \mathbf{DECAY}(2(\Delta t_2 + \Delta t_1)) + \dots + \\
&+ \mathbf{DECAY}((n - 1)(\Delta t_1 + \Delta t_2))] \times \mathbf{DESTR}(\Delta t_1) \times \overline{N}(0) . \tag{16}
\end{aligned}$$

The summation of the  $\mathbf{DECAY}$  matrices in the brackets results in a matrix, which we define as  $\mathbf{SDECAY}$ , where each element can be represented by a combination of geometric series of  $n$  terms of the form

$$1 + e^{-\lambda_i \tau} + e^{-2\lambda_i \tau} + \dots + e^{-(n-1)\lambda_i \tau} = \frac{1 - e^{-n\lambda_i \tau}}{1 - e^{-\lambda_i \tau}}$$

where  $\tau = (\Delta t_1 + \Delta t_2)$ . Equation 16 becomes

$$\bar{N}((n\Delta t_1 + (n-1)\Delta t_2)) = \mathbf{SDECAY} \times \mathbf{DESTR} \times \bar{N}(0). \quad (17)$$

For a three nuclide chain the **SDECAY** matrix exhibits the following form,

$$\mathbf{SDECAY} = \begin{bmatrix} S_1(\tau) & 0.0 & 0.0 \\ \frac{\lambda_1 (S_1(\tau) - S_2(\tau))}{(\lambda_2 - \lambda_1)} & S_2(\tau) & 0.0 \\ \frac{\lambda_1 \lambda_2}{(\lambda_2 - \lambda_1)} \frac{(S_1(\tau) - S_3(\tau))}{(\lambda_3 - \lambda_1)} + \frac{\lambda_2 (S_2(\tau) - S_3(\tau))}{(\lambda_3 - \lambda_2)} & & S_3(\tau) \\ + \frac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)} \frac{(S_2(\tau) - S_3(\tau))}{(\lambda_3 - \lambda_2)} & & \end{bmatrix}.$$

where

$$S_i(\tau) = \frac{1 - e^{-n\lambda_i \tau}}{1 - e^{-\lambda_i \tau}}.$$

Thus by generating the **DESTR** and **SDECAY** matrices and performing matrix multiplication, an exact solution for the case where the initial material concentrations are present at the beginning of each pulse and the transmuted and radioactive nuclide are extracted after passing through the irradiation environment can be obtained. An analytical solution for the first three nuclides within a linear chain is presented in Appendix A. Solutions to more complicated irradiation schedules as depicted in Figure 2 can be generated by grouping common **DECAY** matrices together to form **SDECAY** type matrices which are subsequently multiplied together.

As a side note, the methods currently in use for pulse/intermittent irradiation which preserve the pulsed nature of the irradiation history approximate the **SDECAY** and **DESTR** matrices [7-9]. The **SDECAY** matrix is approximated by its diagonal elements and in several cases the **DESTR** matrix is generated assuming that the parent nuclide is not destroyed during irradiation. These approximations are frequently used to model cases for which Model A is the relevant method. To illustrate the limitations of these models, a comparison of Method B to Method A is presented in Section III.

**Table 1. Fractional Nuclide Concentrations for the  $^{55}_{25}\text{Mn}$  Linear Chain.**

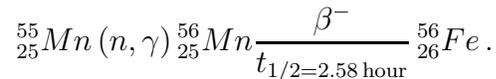
	Number of Pulses 5000			Number of Pulses 250000		
	Analytic	Direct Multiplication	Method A	Analytic	Direct Multiplication	Method A
$N_1(t)$	9.99698(-1)*	9.99698(-1)	9.99698(-1)	9.85020(-1)	9.85020(-1)	9.85020(-1)
$N_2(t)$	9.56048(-8)	9.56048(-8)	9.56048(-8)	9.42011(-8)	9.42011(-8)	9.42011(-8)
$N_3(t)$	3.01719(-4)	3.01719(-4)	3.01719(-4)	1.49796(-2)	1.49796(-2)	1.49796(-2)
Total	1.00000	1.00000	1.00000	1.00000	1.00000	1.00000

\*Read as  $9.99698 \times 10^{-1}$ .

### III Numerical Results

The irradiation history depicted in Fig. 3 which consists of a series of  $n$  uniformly spaced pulses of flux height  $\phi_p$  and various simplified nuclide transmutation/decay chains are used to analyze the pulse/intermittent activation schemes, Methods A and B, of Section II. The destruction and production rates for the nuclide chains were obtained from the radioactivity code DKR-ICF [8] and incorporated into the PULSAR demonstration code [6,10]. EISPACK [27] subroutines were used to determine all eigenvalues and corresponding eigenvectors of a real general matrix. Also, the inverse of  $\mathbf{P}$  was determined using subroutines from LINPACK [28]. These routines were utilized due to their robustness and reliability attained through years of testing and usage.

We begin with the analysis of Method A. One of the tests for any numerical scheme is a comparison to analytical solutions. Table 1 presents a comparison between the results obtained by Method A, the analytical solution given in Appendix A and direct multiplication of the matrices for the uniform series of pulses depicted in Fig. 3. The three nuclide chain analyzed is the formation of  $^{56}_{26}\text{Fe}$  from  $^{55}_{25}\text{Mn}$ . The complete transmutation/decay chain is



The parameters used in the computations are;  $\sigma\phi = 1.677 \times 10^{-11} \text{ s}^{-1}$ , a pulse width of  $\Delta t_1 = 3600 \text{ s}$ , and a dwell time of  $\Delta t_2 = 7200 \text{ s}$ . For simplicity in the analysis, the initial

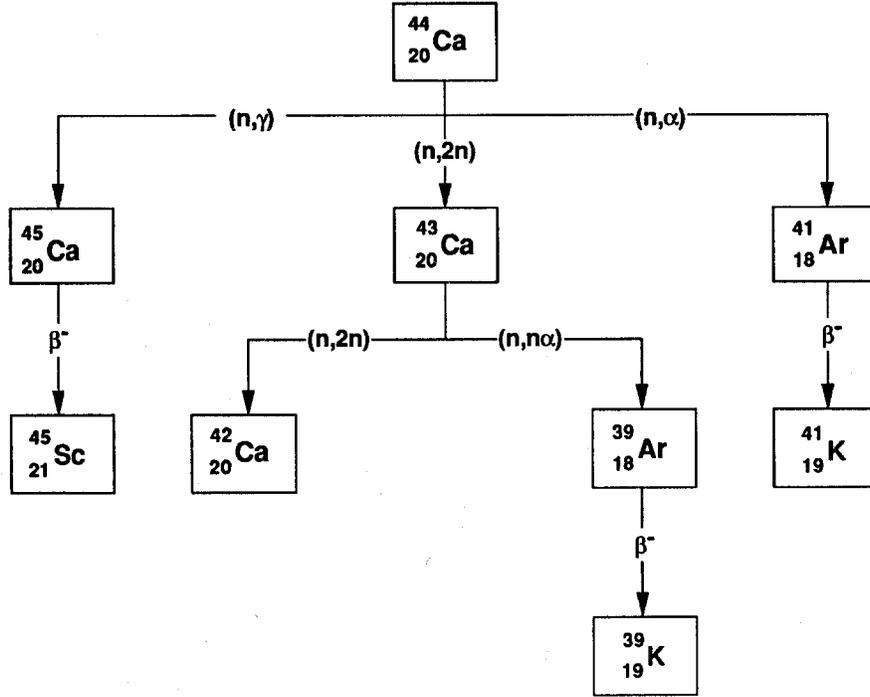
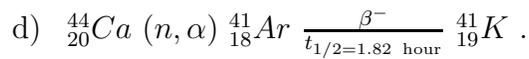
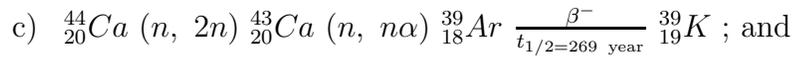
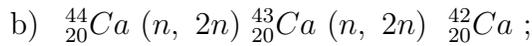
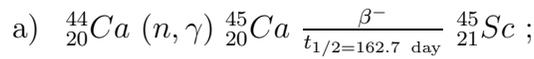


Figure 5.  $^{44}_{20}\text{Ca}$  transmutation and decay scheme.

atomic concentration of  $^{55}_{25}\text{Mn}$  was set to unity. We note that the results agree identically which verifies Method A as well as the analytical solutions.

Figure 5 depicts a simplified transmutation/decay scheme for  $^{44}_{20}\text{Ca}$  which will be analyzed using Method A. The time dependent concentration of the nuclides present in the transmutation/decay scheme can be determined from the following four linear nuclide chains;



**Table 2. Fractional Nuclide Concentrations for the  $^{44}_{20}\text{Ca}$  Transmutation and Decay Scheme**

Nuclide	Case 1 Parameters	Case 2 Parameters
$^{44}_{20}\text{Ca}$ (stable)	9.774 (-1) <sup>a</sup>	9.554 (-1)
$^{45}_{20}\text{Ca}$	2.485 (-4)	6.487 (-4)
$^{45}_{21}\text{Sc}$ (stable)	5.758 (-3)	1.123 (-2)
$^{43}_{20}\text{Ca}$ (stable)	1.409 (-2)	2.765 (-2)
$^{42}_{20}\text{Ca}$ (stable)	9.364 (-5)	3.699 (-4)
$^{41}_{18}\text{Ar}$	6.402 (-8)	1.248 (-7)
$^{41}_{19}\text{K}$ (stable)	2.377 (-3)	4.700 (-3)
$^{39}_{18}\text{Ar}$	1.996 (-4)	9.669 (-5)
$^{39}_{19}\text{K}$ (stable)	2.651 (-6)	9.673 (-7)
Total	1.000	1.000

<sup>a</sup>Read as  $9.774 \times 10^{-1}$ , etc.

Two operation scenerios (irradiation histories) are examined having the following pulse widths (irradiation times),  $\Delta t_1$ , and dwell times,  $\Delta t_2$ ;

- a) Case 1:  $\Delta t_1 = 3600$  s and  $\Delta t_2 = 7200$  s; and
- b) Case 2:  $\Delta t_1 = 3600$  s and  $\Delta t_2 = 900$  s.

The number of pulses considered for both cases is 45,000. The initial atomic concentration of  $^{44}_{20}\text{Ca}$  is set to unity.

The results of Cases 1 and 2 are presented in Table 2. Recall that Method A is exact for the situation where the irradiated material is present throughout a series of pulses. No additions or extractions of the nuclides take place, hence the number of nuclides must be conserved. The results presented in Table 2 indicate that this is indeed so as the summation of all 9 nuclides in the transmutation scheme of Fig. 5 is equal to unity.

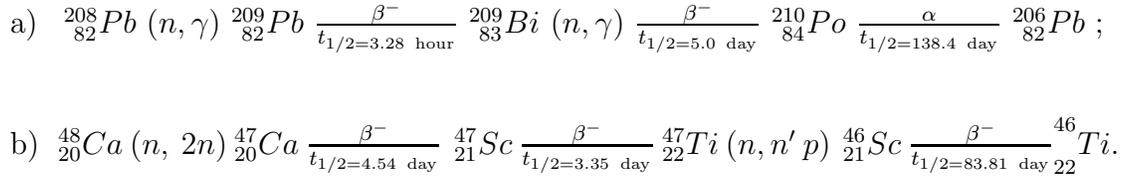
The following two nuclide chains are considered for the analysis of Method B,

**Table 3. Comparison of the Fractional Nuclide Concentrations Computed by Method A and Method B for the  $^{208}_{82}\text{Pb}$  Nuclide Chain.**

Nuclide	45,000 Pulses		285,000 Pulses	
	Method A	Method B	Method A	Method B
$^{208}_{82}\text{Pb}$ (stable)	9.941 (-1) <sup>b</sup>	1.000 <sup>c</sup>	9.634 (-1)	1.000 <sup>c</sup>
$^{209}_{82}\text{Pb}$	1.380 (-8)	1.388 (-8)	1.388 (-8)	1.388 (-8)
$^{209}_{83}\text{Bi}$ (stable)	3.232 (-4)	3.254 (-4)	1.974 (-3)	2.061 (-3)
$^{210}_{83}\text{Bi}$	2.033 (-10)	1.525 (-16)	1.243 (-9)	1.525 (-16)
$^{210}_{84}\text{Po}$	1.560 (-9)	4.184 (-15)	9.618 (-9)	4.184 (-15)
$^{206}_{82}\text{Pb}$ (stable)	2.178 (-8)	1.154 (-13)	8.692 (-7)	7.539 (-13)
Total	9.944 (-1)	1.0003	9.654 (-1)	1.002

<sup>b</sup>Read as  $9.941 \times 10^{-1}$ , etc.

<sup>c</sup>Has been rounded to 1.000



Two operation scenerios, one consisting of 45,000 pulses and the other of 285,000 pulses both having pulse widths of 3600 s and dwell times of 7200 s are examined.

We recall that Method B assumes that the initial parent nuclide concentration is present at the beginning of each pulse and that the transmuted and radioactive nuclides are extracted between pulses. Recall also that approximate solution schemes to Method B (Section II.B.2) [7-9] are used to model the situation where the irradiated material is present throughout the series of pulses (the case for which Method A is exact). A comparison of both methods will indicate where Method B breaks down in its approximation of Method A and hence where these approximate solution schemes yield incorrect results.

**Table 4. Comparison of the Fractional Nuclide Concentrations Computed by Method A and Method B for the  $^{48}_{20}\text{Ca}$  Nuclide Chain.**

Nuclide	45,000 Pulses		285,000 Pulses	
	Method A	Method B	Method A	Method B
$^{48}_{20}\text{Ca}$ (stable)	9.707 (-1) <sup>c</sup>	1.000 <sup>d</sup>	8.281 (-1)	1.000 <sup>d</sup>
$^{47}_{20}\text{Ca}$	2.780 (-5)	2.372 (-5)	2.372 (-5)	2.864 (-5)
$^{47}_{21}\text{Sc}$	2.085 (-5)	2.148 (-5)	1.779 (-5)	2.148 (-5)
$^{47}_{22}\text{Ti}$ (stable)	2.373 (-2)	2.443 (-2)	1.291 (-1)	1.550 (-1)
$^{46}_{21}\text{Sc}$	6.714 (-8)	1.975 (-18)	3.720 (-7)	1.975 (-18)
$^{46}_{22}\text{Ti}$ (stable)	1.541 (-6)	3.778 (-14)	5.808 (-5)	2.393 (-13)
Total	9.944 (-1)	1.024	9.573 (-1)	1.155

<sup>c</sup>Read as  $9.707 \times 10^{-1}$ , etc.

<sup>d</sup>Has been rounded to 1.000.

Table 3 presents a comparison of the 45,000 and 285,000 pulse results computed by Methods A and B for the  $^{208}_{82}\text{Pb}$  nuclide chain. First, we note that though the  $^{209}_{83}\text{Bi}$  concentration (a stable nuclide) of Method B compares well with that of Method A, the concentrations of the nuclides following  $^{209}_{83}\text{Bi}$  are in complete disagreement, differing by orders of magnitude. Second, the summation of all nuclides in the chain is larger for Method B than for Method A. This is because Method B assumes that the initial parent nuclide concentration is present at the beginning of each pulse. In effect, we are adding a small amount of parent nuclide at each pulse. (Note: Method A does not sum to unity as only one linear nuclide chain from the  $^{208}_{82}\text{Pb}$  transmutation/decay scheme was selected. The fraction missing is the contribution from other chains in the transmutation/decay scheme.)

Table 4 presents the comparison of Methods A and B for 45,000 and 285,000 pulses for the  $^{48}_{20}\text{Ca}$  nuclide chain. As with the  $^{208}_{82}\text{Pb}$  chain, we note a complete disagreement in Method B versus Method A results following the first stable descendant in the chain. The radioactive nuclides preceding the stable descendant compare quite favorably especially for the low pulse case (low fluence case) of 45,000 pulses. Once again we note that the sum of the nuclide concentrations for Method B exceeds that of Method A.

From the above comparison we conclude that Method B is a good to fair approximation to Method A provided the total fluence is small and for radioactive nuclides preceding a stable descendant in the chain. It completely breaks down for nuclides following a stable descendant in the chain. Hence care must be exercised in applying Method B for situations where Method A is the appropriate model to use, particularly if one is interested in a radionuclide which follows a stable daughter in a transmutation/decay scheme. This conclusion applies to those codes which utilize the approximate solution schemes indicated in Section II.B.2 as Method B is an improvement to these methods.

## IV Summary

Two calculational methods have been developed to compute the induced activity due to pulsed/intermittent irradiation histories as encountered in both magnetic and inertial fusion energy devices. The numerical solution algorithms are based on the linear chain method and employ series reduction and matrix algebra. The first method (Method A) models the case in which the irradiated materials are further transmuted during each pulse and applies to any structural material or component which resides in the irradiation environment for an extended period of time. No constant production or destruction rate assumptions are made. The second method (Method B) treats the case where a fixed amount of radioactive and transmuted material is created during each pulse. Constant production and destruction rates are assumed for each pulse. This method is exact for the case where the transmuted and activated materials are extracted after each pulse. The two calculational methods can be combined to model irradiation histories that have characteristics of each method. Replacement of a sensitive material or diagnostic component on a periodic basis due to radiation damage in the irradiation environment would require the use of a computational scheme employing both methods.

The efficiency of the numerical scheme for the evaluation of Method A is the result of the lower triangularity of the Bateman Equation solution matrix, the lower triangularity of the **DECAY-DESTR** product matrix and the existence of an efficient matrix decomposition method for raising a matrix to a power.

In Section II.B.2 it was noted that approximations to Method B, when employed to model situations for which Method A is the appropriate method, tend to yield good results for short operating times and for radioactive daughters and can lead to significant errors for long operating times and for nuclides beyond a stable descendant in a chain. Specifically, nuclides following a stable descendant in a chain differed by several orders of magnitude from the correct solution. The comparison between Methods A and B in

the results section clearly illustrated this. This can have dire consequences if the nuclide in question is hazardous.

Though these computational methods have been developed for use in fusion energy systems, the application to fusion systems is not unique. One can use these methods for any application where materials are intermittently exposed to an irradiation environment. Two examples are recirculating fluids in a fission reactor and exposure of materials in beam dumps of high energy accelerators.

## Appendix A: Analytical Results

Analytical solutions for a three nuclide chain are presented for both methods outlined in Section II. As can be noted from the solutions, the complexity of the solutions increases as one proceeds to nuclides further down a linear chain.

The analytical solution for Method A, the case where the materials are present throughout a series of irradiation pulses, is:

$$N_1(t) = N_1(0) e^{-nd_1\Delta t_1} e^{-(n-1)\lambda_1\Delta t_2} \quad (18)$$

$$\begin{aligned} N_2(t) &= N_2(0) e^{-nd_2\Delta t_1} e^{-(n-1)\lambda_2\Delta t_2} \\ &+ \lambda_1 N_1(0) \left( \frac{e^{-\lambda_1\Delta t_2} - e^{-\lambda_2\Delta t_2}}{\lambda_2 - \lambda_1} \right) e^{-d_1\Delta t_1} e^{-d_2\Delta t_1} \times \\ &\quad \times \left( \frac{e^{-(n-1)(d_2\Delta t_1 + \lambda_2\Delta t_2)} - e^{-(n-1)(d_1\Delta t_1 + \lambda_1\Delta t_2)}}{e^{-(d_2\Delta t_1 + \lambda_2\Delta t_2)} - e^{-(d_1\Delta t_1 + \lambda_1\Delta t_2)}} \right) \\ &+ P_2 N_1(0) \left( \frac{e^{-d_1\Delta t_1} - e^{-d_2\Delta t_1}}{d_2 - d_1} \right) \times \\ &\quad \times \left( \frac{e^{-n(d_2\Delta t_1 + \lambda_2\Delta t_2)} - e^{-n(d_1\Delta t_1 + \lambda_1\Delta t_2)}}{e^{-(d_2\Delta t_1 + \lambda_2\Delta t_2)} - e^{-(d_1\Delta t_1 + \lambda_1\Delta t_2)}} \right) \end{aligned} \quad (19)$$

$$\begin{aligned} N_3(t) &= N_3(0) e^{-nd_3\Delta t_1} e^{-(n-1)\lambda_3\Delta t_2} \\ &+ P_3 N_2(0) \left( \frac{e^{-d_2\Delta t_1} - e^{-d_3\Delta t_1}}{d_3 - d_2} \right) \times \\ &\quad \times \left( \frac{e^{-n(d_2\Delta t_1 + \lambda_2\Delta t_2)} - e^{-n(d_3\Delta t_1 + \lambda_3\Delta t_2)}}{e^{-(d_2\Delta t_1 + \lambda_2\Delta t_2)} - e^{-(d_3\Delta t_1 + \lambda_3\Delta t_2)}} \right) \\ &+ \lambda_2 N_2(0) \left( \frac{e^{-\lambda_2\Delta t_2} - e^{-\lambda_3\Delta t_2}}{\lambda_3 - \lambda_2} \right) e^{-d_2\Delta t_1} e^{-d_3\Delta t_1} \times \\ &\quad \times \left( \frac{e^{-(n-1)(d_2\Delta t_1 + \lambda_2\Delta t_2)} - e^{-(n-1)(d_3\Delta t_1 + \lambda_3\Delta t_2)}}{e^{-(d_2\Delta t_1 + \lambda_2\Delta t_2)} - e^{-(d_3\Delta t_1 + \lambda_3\Delta t_2)}} \right) \\ &+ P_2 P_3 N_1(0) \left( \frac{e^{-d_1\Delta t_1} - e^{-d_3\Delta t_1}}{(d_2 - d_1)(d_3 - d_1)} + \frac{e^{-d_2\Delta t_1} - e^{-d_3\Delta t_1}}{(d_1 - d_2)(d_3 - d_2)} \right) \times \\ &\quad \times \left( \frac{e^{-n(d_1\Delta t_1 + \lambda_1\Delta t_2)} - e^{-n(d_3\Delta t_1 + \lambda_3\Delta t_2)}}{e^{-(d_1\Delta t_1 + \lambda_1\Delta t_2)} - e^{-(d_3\Delta t_1 + \lambda_3\Delta t_2)}} \right) \\ &+ \lambda_1 N_1(0) \left( \frac{e^{-\lambda_1\Delta t_2} - e^{-\lambda_2\Delta t_2}}{\lambda_2 - \lambda_1} \right) P_3 \left( \frac{e^{-d_2\Delta t_1} - e^{-d_3\Delta t_1}}{d_3 - d_2} \right) e^{-d_1\Delta t_1} \times \\ &\quad \times \left( \frac{e^{-(n-1)(d_1\Delta t_1 + \lambda_1\Delta t_2)} - e^{-(n-1)(d_3\Delta t_1 + \lambda_3\Delta t_2)}}{e^{-(d_1\Delta t_1 + \lambda_1\Delta t_2)} - e^{-(d_3\Delta t_1 + \lambda_3\Delta t_2)}} \right) \end{aligned}$$

$$\begin{aligned}
& + \lambda_1 \lambda_2 N_1(0) \left( \frac{e^{-\lambda_1 \Delta t_2} - e^{-\lambda_3 \Delta t_2}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} + \frac{e^{-\lambda_2 \Delta t_2} - e^{-\lambda_3 \Delta t_2}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} \right) \times \\
& \quad \times e^{-d_1 \Delta t_1} e^{-d_3 \Delta t_1} \left( \frac{e^{-(n-1)(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} - e^{-(n-1)(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}}{e^{-(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} - e^{-(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}} \right) \\
& + N_1(0) \left( P_2 \left( \frac{e^{-d_1 \Delta t_1} - e^{-d_2 \Delta t_1}}{d_2 - d_1} \right) e^{-(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} + \right. \\
& \quad \left. + \lambda_1 \left( \frac{e^{-\lambda_1 \Delta t_2} - e^{-\lambda_2 \Delta t_2}}{\lambda_2 - \lambda_1} \right) e^{-d_1 \Delta t_1} e^{-d_2 \Delta t_1} \right) \times \\
& \quad \times \left( P_3 e^{-\lambda_2 \Delta t_2} \left( \frac{e^{-d_2 \Delta t_1} - e^{-d_3 \Delta t_1}}{d_3 - d_2} \right) + \lambda_2 e^{-d_3 \Delta t_1} \left( \frac{e^{-\lambda_2 \Delta t_2} - e^{-\lambda_3 \Delta t_2}}{\lambda_3 - \lambda_2} \right) \right) \times \\
& \quad \times \left( \frac{e^{-(n-1)(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} - e^{-(n-1)(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}}{(e^{-(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} - e^{-(d_3 \Delta t_1 + \lambda_3 \Delta t_2)})(e^{-(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} - e^{-(d_2 \Delta t_1 + \lambda_2 \Delta t_2)})} \right) \\
& + N_1(0) \left( P_2 \left( \frac{e^{-d_1 \Delta t_1} - e^{-d_2 \Delta t_1}}{d_2 - d_1} \right) e^{-(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} + \right. \\
& \quad \left. + \lambda_1 \left( \frac{e^{-\lambda_1 \Delta t_2} - e^{-\lambda_2 \Delta t_2}}{\lambda_2 - \lambda_1} \right) e^{-d_1 \Delta t_1} e^{-d_2 \Delta t_1} \right) \times \\
& \quad \times \left( P_3 e^{-\lambda_2 \Delta t_2} \left( \frac{e^{-d_2 \Delta t_1} - e^{-d_3 \Delta t_1}}{d_3 - d_2} \right) + \lambda_2 e^{-d_3 \Delta t_1} \left( \frac{e^{-\lambda_2 \Delta t_2} - e^{-\lambda_3 \Delta t_2}}{\lambda_3 - \lambda_2} \right) \right) \times \\
& \quad \times \left( \frac{e^{-(n-1)(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-(n-1)(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}}{(e^{-(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-(d_3 \Delta t_1 + \lambda_3 \Delta t_2)})(e^{-(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-(d_1 \Delta t_1 + \lambda_1 \Delta t_2)})} \right)
\end{aligned} \tag{20}$$

Note here that in addition to neutron destruction, the parent nuclide  $N_1$  is assumed to be radioactive. If  $N_1$  is a stable nuclide, the above solutions reduce to

$$N_1(t) = N_1(0) e^{-nd_1 \Delta t_1} \tag{21}$$

$$\begin{aligned}
N_2(t) & = N_2(0) e^{-nd_2 \Delta t_1} e^{-(n-1)\lambda_2 \Delta t_2} \\
& + P_2 N_1(0) \left( \frac{e^{-d_1 \Delta t_1} - e^{-d_2 \Delta t_1}}{d_2 - d_1} \right) \times \\
& \quad \times \left( \frac{e^{-n(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-nd_1 \Delta t_1}}{e^{-(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-d_1 \Delta t_1}} \right)
\end{aligned} \tag{22}$$

$$\begin{aligned}
N_3(t) & = N_3(0) e^{-nd_3 \Delta t_1} e^{-(n-1)\lambda_3 \Delta t_2} \\
& + P_3 N_2(0) \left( \frac{e^{-d_2 \Delta t_1} - e^{-d_3 \Delta t_1}}{d_3 - d_2} \right) \times \\
& \quad \times \left( \frac{e^{-n(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-n(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}}{e^{-(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}} \right)
\end{aligned}$$

$$\begin{aligned}
& + \lambda_2 N_2(0) \left( \frac{e^{-\lambda_2 \Delta t_2} - e^{-\lambda_3 \Delta t_2}}{\lambda_3 - \lambda_2} \right) e^{-d_2 \Delta t_1} e^{-d_3 \Delta t_1} \times \\
& \quad \times \left( \frac{e^{-(n-1)(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-(n-1)(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}}{e^{-(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}} \right) \\
& + P_2 P_3 N_1(0) \left( \frac{e^{-d_1 \Delta t_1} - e^{-d_3 \Delta t_1}}{(d_2 - d_1)(d_3 - d_1)} + \frac{e^{-d_2 \Delta t_1} - e^{-d_3 \Delta t_1}}{(d_1 - d_2)(d_3 - d_2)} \right) \times \\
& \quad \times \left( \frac{e^{-n(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} - e^{-n(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}}{e^{-(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} - e^{-(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}} \right) \\
& + N_1(0) \left( P_2 \left( \frac{e^{-d_1 \Delta t_1} - e^{-d_2 \Delta t_1}}{d_2 - d_1} \right) e^{-(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} \times \right. \\
& \quad \times \left( P_3 e^{-\lambda_2 \Delta t_2} \left( \frac{e^{-d_2 \Delta t_1} - e^{-d_3 \Delta t_1}}{d_3 - d_2} \right) + \lambda_2 e^{-d_3 \Delta t_1} \left( \frac{e^{-\lambda_2 \Delta t_2} - e^{-\lambda_3 \Delta t_2}}{\lambda_3 - \lambda_2} \right) \right) \times \\
& \quad \times \left. \left( \frac{e^{-(n-1)(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} - e^{-(n-1)(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}}{(e^{-(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} - e^{-(d_3 \Delta t_1 + \lambda_3 \Delta t_2)})(e^{-(d_1 \Delta t_1 + \lambda_1 \Delta t_2)} - e^{-(d_2 \Delta t_1 + \lambda_2 \Delta t_2)})} \right) \right) \\
& + N_1(0) \left( P_2 \left( \frac{e^{-d_1 \Delta t_1} - e^{-d_2 \Delta t_1}}{d_2 - d_1} \right) e^{-(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} \times \right. \\
& \quad \times \left( P_3 e^{-\lambda_2 \Delta t_2} \left( \frac{e^{-d_2 \Delta t_1} - e^{-d_3 \Delta t_1}}{d_3 - d_2} \right) + \lambda_2 e^{-d_3 \Delta t_1} \left( \frac{e^{-\lambda_2 \Delta t_2} - e^{-\lambda_3 \Delta t_2}}{\lambda_3 - \lambda_2} \right) \right) \times \\
& \quad \times \left. \left( \frac{e^{-(n-1)(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-(n-1)(d_3 \Delta t_1 + \lambda_3 \Delta t_2)}}{(e^{-(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-(d_3 \Delta t_1 + \lambda_3 \Delta t_2)})(e^{-(d_2 \Delta t_1 + \lambda_2 \Delta t_2)} - e^{-(d_1 \Delta t_1 + \lambda_1 \Delta t_2)})} \right) \right)
\end{aligned} \tag{23}$$

where  $d_1$  is now  $\sigma_1 \Phi$ .

The analytical solution for Method B, the case where the initial material concentrations are present at the beginning of each pulse and the transmuted and radioactive nuclides are extracted after passing through the irradiation environment, is:

$$N_1(t) = N_1(0) e^{-d_1 \Delta t_1} \left( \frac{1 - e^{-n \lambda_1 \tau}}{1 - e^{-\lambda_1 \tau}} \right) \tag{24}$$

$$\begin{aligned}
N_2(t) & = N_2(0) e^{-d_2 \Delta t_1} \left( \frac{1 - e^{-n \lambda_2 \tau}}{1 - e^{-\lambda_2 \tau}} \right) \\
& + P_2 N_1(0) \left( \frac{e^{-d_1 \Delta t_1} - e^{-d_2 \Delta t_1}}{d_2 - d_1} \right) \left( \frac{1 - e^{-n \lambda_2 \tau}}{1 - e^{-\lambda_2 \tau}} \right) \\
& + N_1(0) e^{-d_1 \Delta t_1} \frac{\lambda_1}{\lambda_2 - \lambda_1} \left( \frac{1 - e^{-n \lambda_1 \tau}}{1 - e^{-\lambda_1 \tau}} - \frac{1 - e^{-n \lambda_2 \tau}}{1 - e^{-\lambda_2 \tau}} \right)
\end{aligned} \tag{25}$$

$$\begin{aligned}
N_3(t) = & N_3(0) e^{-d_3 \Delta t_1} \left( \frac{1 - e^{-n \lambda_3 \tau}}{1 - e^{-\lambda_3 \tau}} \right) \\
& + P_3 N_2(0) \left( \frac{e^{-d_2 \Delta t_1} - e^{-d_3 \Delta t_1}}{d_3 - d_2} \right) \left( \frac{1 - e^{-n \lambda_3 \tau}}{1 - e^{-\lambda_3 \tau}} \right) \\
& + N_2(0) e^{-d_2 \Delta t_1} \frac{\lambda_2}{\lambda_3 - \lambda_2} \left( \frac{1 - e^{-n \lambda_2 \tau}}{1 - e^{-\lambda_2 \tau}} - \frac{1 - e^{-n \lambda_3 \tau}}{1 - e^{-\lambda_3 \tau}} \right) \\
& + P_2 P_3 N_1(0) \left( \frac{e^{-d_1 \Delta t_1} - e^{-d_3 \Delta t_1}}{(d_2 - d_1)(d_3 - d_1)} + \frac{e^{-d_2 \Delta t_1} - e^{-d_3 \Delta t_1}}{(d_1 - d_2)(d_3 - d_2)} \right) \left( \frac{1 - e^{-n \lambda_3 \tau}}{1 - e^{-\lambda_3 \tau}} \right) \\
& + P_2 N_1(0) \left( \frac{e^{-d_1 \Delta t_1} - e^{-d_2 \Delta t_1}}{d_2 - d_1} \right) \frac{\lambda_2}{\lambda_3 - \lambda_2} \left( \frac{1 - e^{-n \lambda_2 \tau}}{1 - e^{-\lambda_2 \tau}} - \frac{1 - e^{-n \lambda_3 \tau}}{1 - e^{-\lambda_3 \tau}} \right) \\
& + N_1(0) e^{-d_1 \Delta t_1} \left( \frac{\lambda_1 \lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} \left( \frac{1 - e^{-n \lambda_1 \tau}}{1 - e^{-\lambda_1 \tau}} - \frac{1 - e^{-n \lambda_2 \tau}}{1 - e^{-\lambda_2 \tau}} \right) + \right. \\
& \left. + \frac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} \left( \frac{1 - e^{-n \lambda_2 \tau}}{1 - e^{-\lambda_2 \tau}} - \frac{1 - e^{-n \lambda_3 \tau}}{1 - e^{-\lambda_3 \tau}} \right) \right) \tag{26}
\end{aligned}$$

where

$$\begin{aligned}
P_2 &= \text{neutron production rate of nuclide 2} = \sigma_1 \Phi, \text{ or } \lambda_1; \\
P_3 &= \text{neutron production rate of nuclide 3} = \sigma_2 \Phi, \text{ or } \lambda_2; \\
d_1 &= \text{destruction rate of nuclide 1} = \sigma_1 \Phi + \lambda_1; \\
d_2 &= \text{destruction rate of nuclide 2} = \sigma_2 \Phi + \lambda_2; \\
d_3 &= \text{destruction rate of nuclide 3} = \sigma_3 \Phi + \lambda_3; \\
\Delta_1 &= \text{the pulse width;} \\
\Delta_2 &= \text{the dwell (off) time;} \\
\tau &= \Delta_1 + \Delta_2 \quad .
\end{aligned}$$

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**Calculational Models for the Treatment of  
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