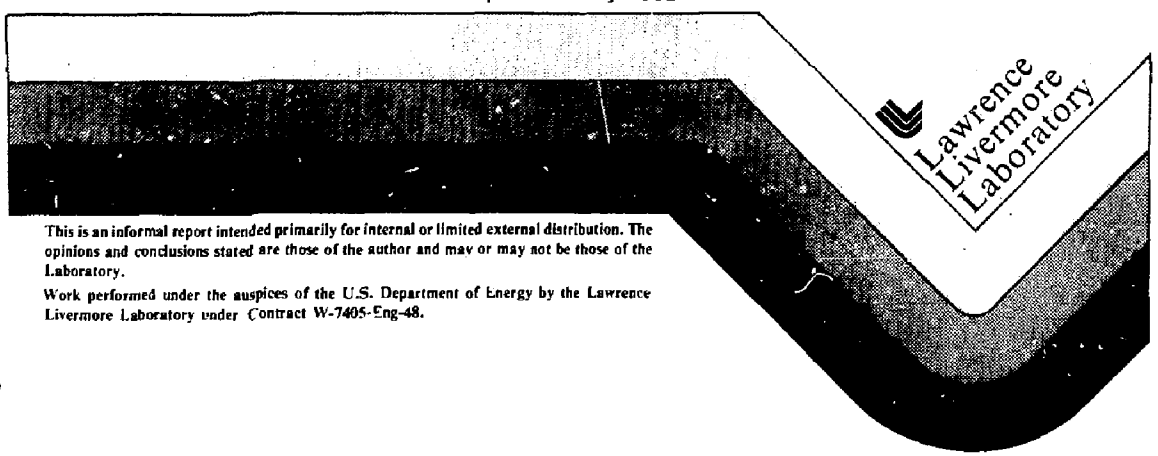# GENERAL FORMALISM FOR THE STUDY OF ACTIVATION: APPLICATION TO RADIOCHEMICAL DETECTORS

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General Formalism for the Study of Activation:

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Abstract

This paper develops mathematical techniques required for the
study of neutron-induced activation of importance to fission
and fusion devices -- reactors, nuclear weapons, etc. The
formalism is presented as a guide for examining the dependence
of activation products on flux time history, spatial gradients
and the sensitivity to the assumed reactions and cross
sections. Exact solutions in powers of the neutron fluence
are presented and examined in various limits. As an example,
radiochemical threshold (n,2n) detectors used to diagnose
thermonuclear explosions are studied using approximations to
these solutions. In particular, approximate formulas for the
sensitivity of the radiochemical products to different cross
sections are derived.

I.   INTRODUCTION

Large neutron fluences are produced in fission power and research reactors, in nuclear weapons and in neutron stars and other astrophysical phenomena. In addition, fusion reactors burning deuterium or tritium fuel will also produce large quantities of neutrons. Activation of materials is a major consequence of these neutrons and may be a benefit or a problem depending on the circumstances. For example, the radioactive products from special detector materials loaded in a thermonuclear device are important measures of device performance.[1] In a power reactor, however, certain radioactive products present a significant waste disposal hazard and are to be avoided.

The calculation of the amount of any radioactive product produced by a given neutron fluence is, in principal, straightforward, but may, in fact, be complicated by the presence of strong energy and time dependences in the neutron flux, lack of spatial homogeneity in both the material and the neutron field, the occurrence of multiple reactions and the uncertainty of certain cross sections. Most of these difficulties may be handled by suitable computer codes, but these codes often obscure relationships which may be important to gain a better understanding of particular problems. An example of this is understanding the sensitivity of the production of certain isotopes to various assumed intermediate steps and their cross sections. In a thermonuclear device, for example, isotopes several mass units from the stable loaded material can be produced by multiple (n,2n) reactions. If such products are used for diagnostics, it is important to know, for

example, how neutron capture on one of the intermediate isotopes affects the final answer. Although such a problem can be studied by activation codes, a general formalism is needed to guide such studies, to identify correlations and other relationships, and to focus attention on important parameters. In this paper, such a formalism is developed and applied in a general way to the radiochemical diagnosis of thermonuclear devices. In a companion paper[2] (classified) the formalism is applied to some specific problems for threshold $(n,2n)$ detectors.

II. PROBLEM STATEMENT

In general, we wish to consider a medium in which is present a time-varying, energy-dependent neutron flux with spatial gradients. In this medium we consider the production and destruction of a number K of isotopes and isomers and require the amount $N_j(\tau)$ of the $j^{\underline{th}}$ isotope or isomer $(j=1,\ldots,K)$ at time τ. The initial population is $N_j(0) \equiv N_{0j}$. The different members of the set are connected by neutron-induced cross sections, $\sigma_{ij}(E)$, where i (j) refers to the final (initial) isotope or isomer and E is the neutron energy. Cross sections on a species j which do not have as their residual nucleus another member of the set K are combined into a single quantity $\sigma_{Dj}(E)$ (D stands for destruction). In addition, members of the set may decay spontaneously; this is characterized by the decay constant $\lambda_j = T_j^{-1}$, where T_j is the mean life of species j. The decay constant λ_j is the sum of all partial decay constants λ_{ij} leading to other species i within the set K and λ_{0j} leading to residual nuclei not included within the set.

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To begin with, we will consider a portion of the medium, called a zone, small enough that both the neutron flux and the population $N(\tau)$ can be considered to be uniform within it. We will first derive the problem for a single zone. Within the zone let $\varphi(\tau, E)dE$ be the instantaneous number of neutrons per unit area per unit time with energies between E and E+dE. This will be referred to as the flux spectrum and

$$\varphi(\tau) = \int_0^\infty \varphi(\tau, E)dE$$

is the flux at time τ. The total neutron fluence after an elapsed time t is given as:

$$\Phi(t) = \int_0^t \varphi(\tau)d\tau$$

and one may define a fluence spectrum

$$\Phi(t, E) = \int_0^t \varphi(\tau, E)d\tau$$

or other special fluences

$$\Phi(t, \Delta E) = \int_{\Delta E} \Phi(t, E)dE$$

where, for example, ΔE may be a region of the neutron spectrum around 14 MeV. The latter quantity, then, is referred to as the "14-MeV fluence."

For species i, the increase in its population during interval $d\tau$, because of neutron-induced reactions, from other members in the set is

$$\sum_{\substack{j=1 \\ (j \neq i)}}^{K} [\int_{0}^{\infty} \sigma_{ij}(E) \, \varphi \, (\tau, E) dE \,] \, N_j(\tau) d\tau$$

and the increase as the result of decays of other members to species i is

$$\sum_{\substack{j=1 \\ (j \neq i)}}^{K} \lambda_{ij} \, N_j(\tau) d\tau \quad .$$

During the same time interval, the decrease in population of species i from neutron-induced reactions on it is

$$- [\sum_{k \neq i}^{K} \int_{0}^{\infty} \sigma_{ki}(E) \, \varphi \, (\tau, E) dE \, + \, \int_{0}^{\infty} \sigma_{Di}(E) \, \varphi \, (\tau, E) dE \,] \, N_i(\tau) d\tau$$

and the decrease from spontaneous decay is $- \lambda_i \, N_i \, d\tau$. These relations may be combined to give the total change in the population of species i in time $d\tau$, namely

$$\frac{dN_i(\tau)}{d\tau} = \sum_{j=1}^{K} [\int_{0}^{\infty} \sigma_{ij}(E) \varphi(\tau, E) dE \, + \, a_{ij} \lambda_j] \, N_j(\tau) \quad . \tag{1}$$

In Eq. 1 σ_{ii} is defined as

$$\sigma_{ii} = - \sum_{k \neq i}^{K} \sigma_{ki} - c_{Di} \tag{2}$$

and a_{ij} is the branching ratio λ_{ij}/λ_j for species j with a_{ii} defined as -1. Equation 1 may be written in matrix notation

$$\frac{d\vec{N}}{d\tau} = [\int_0^\infty \underset{\sim}{\sigma}(E)\,\varphi\,(\tau,E)dE + \underset{\sim}{a} \cdot \underset{\sim}{\lambda}\,] \cdot \vec{N} \qquad (3)$$

where $\vec{N}$ is a vector whose elements are the instantaneous populations of the members of set K and $\underset{\sim}{\sigma}(E)$ is a matrix whose off-diagonal elements are the energy-dependent coupling cross sections $\sigma_{ij}(E)$ and whose diagonal elements are defined in Eq. 2 above. $\underset{\sim}{\lambda}$ is a diagonal matrix whose elements are the total decay constants of each species and $\underset{\sim}{a}$ is a matrix of branching ratios with diagonal elements equal to -1. For a given set K, a given set of cross sections, branching ratios and decay constants, the problem is to solve Eq. 3 in each zone for the neutron flux in that zone. We will examine Eq. 3 in various limits and with different assumptions.

III.   PROBLEM SOLUTION

    1.   Formal Solution by Matrix Diagonalization

        In Eq. 3 the quantity in brackets may be considered to be a matrix operating on $\vec{N}$. This matrix has a time-dependent part and a time-independent part. The solution by diagonalization is appropriate if this operator is not a function of the independent variable. We will examine some circumstances for which this is the case approximately or exactly and write Eq. 3 as

$$\frac{d\vec{N}}{dx} = \underset{\sim}{B} \cdot \vec{N} \qquad (4)$$

where x is the independent variable (τ in Eq. 3) and $\underset{\sim}{B}$ is not a function of x.

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Let $\underset{\sim}{A}$ be a matrix which diagonalizes[3] $\underset{\sim}{B}$, i.e.

$$\underset{\sim}{\gamma} = \underset{\sim}{A}^{-1} \cdot \underset{\sim}{B} \cdot \underset{\sim}{A}$$

where $\gamma_{ij} = \gamma_i \delta_{ij}$. Consequently, for $\underset{\sim}{A} \neq \underset{\sim}{A}(x)$, we may

multiply Eq. 4 on the left by $\underset{\sim}{A}^{-1}$ and using

$\underset{\sim}{A} \cdot \underset{\sim}{A}^{-1} = \underset{\sim}{A}^{-1} \cdot \underset{\sim}{A} = \underset{\sim}{1}$, rewrite it as

$$\underset{\sim}{A}^{-1} \cdot \frac{d\vec{N}}{dx} = \frac{d}{dx}\left(\underset{\sim}{A}^{-1} \cdot \vec{N}\right) = \underset{\sim}{A}^{-1} \cdot \underset{\sim}{B} \cdot \underset{\sim}{A} \cdot \underset{\sim}{A}^{-1} \cdot \vec{N}$$

which becomes

$$\frac{d\vec{Y}}{dx} = \underset{\sim}{\gamma} \cdot \vec{Y} \tag{5}$$

where $\vec{N} = \underset{\sim}{A} \cdot \vec{Y}$ and $\underset{\sim}{\gamma}$ is diagonal. The set of coupled

equations (4) are now uncoupled, the individual elements have

solutions $Y_i = e^{\gamma_i x}$, and the desired populations N_i

are given simply by

$$N_i = \sum_{j=1}^{K} A_{ij} e^{\gamma_j x}$$

where the A_{ij} are determined by diagonalizing $\underset{\sim}{B}$ and γ_i are

the eigenvalues. The normalization of $\underset{\sim}{A}$ is determined by the

initial conditions $N_i(0) = \sum_{j=1}^{K} A_{ij}$.

A.   Example - radioactive chain: Figure 1 illustrates a

typical radioactive chain, with one nuclide feeding the next.

We assume that only N_1 is present at $\tau=0$ and desire the

number of any of the species at time τ later. In the absence

of a neutron flux, the operator $\underset{\sim}{B} = \underset{\sim}{a} \cdot \underset{\sim}{\lambda}$ is independent of

τ. For the case illustrated, and further assuming that the

decays do not branch to any other states, the matrix $\underset{\sim}{a}$ has the

form

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$$
\underset{\sim}{a} = \begin{pmatrix} -1 & 0 & 0 & \cdots & 0 & 0 \\ 1 & -1 & 0 & \cdots & 0 & 0 \\ 0 & 1 & -1 & \cdots & 0 & 0 \\ 0 & 0 & 1 & \cdots & 0 & 0 \\ \cdot & \cdot & \cdot & & \cdot & \cdot \\ \cdot & \cdot & \cdot & & \cdot & \cdot \\ \cdot & \cdot & \cdot & & \cdot & \cdot \\ 0 & 0 & 0 & \cdots & 1 & -1 \end{pmatrix}
$$

The eigenvalues of $\underset{\sim}{a} \cdot \underset{\sim}{\lambda}$ are simply the negative of the decay constants λ_i and the solution for any N_i is simply

$$
N_i = \sum_{j=1}^{K} A_{ij} e^{-\lambda_j \tau}
$$

where the A_{ij} are determined from

$$
-A_{ik}\lambda_k = \sum_j (\underset{\sim}{a}\cdot\underset{\sim}{\lambda})_{ij} A_{jk} \quad . \tag{6}
$$

Using the structure of $\underset{\sim}{a}$, Eq. 6 leads to the recurrence relation,

$$
A_{ik} = \frac{\lambda_{i-1}}{\lambda_i - \lambda_k} A_{i-1,k} \quad , \tag{7}
$$

from which it follows that $A_{ik} = 0$ for $i < k$ and A_{ii} is undetermined ($\lambda_0 \equiv 0$). Equation 7 then yields the set

$$
A_{k1} = \frac{\lambda_1}{\lambda_2 - \lambda_1} \quad \frac{\lambda_2}{\lambda_3 - \lambda_1} \cdots \frac{\lambda_{k-1}}{\lambda_k - \lambda_1} A_{11}
$$

$$
A_{k2} = \frac{\lambda_2}{\lambda_3 - \lambda_2} \quad \frac{\lambda_3}{\lambda_4 - \lambda_2} \cdots \frac{\lambda_{k-1}}{\lambda_k - \lambda_2} A_{22} \tag{8}
$$

$$
A_{k3} = \frac{\lambda_3}{\lambda_4 - \lambda_3} \quad \frac{\lambda_4}{\lambda_5 - \lambda_3} \cdots \frac{\lambda_{k-1}}{\lambda_k - \lambda_3} A_{33}
$$

.
.

etc.

The coefficients A_{ii} are derived from the normalization conditions

$$N_1(0) = N_0 = \sum_{j=1}^{K} A_{1j} \quad \text{and} \quad 0 = \sum_{j=1}^{K} A_{ij} \quad , \quad i \neq 1 \quad (9)$$

Using the fact that $A_{i<j} = 0$ and Eqs. 8 and 9, it follows that

$$
\begin{aligned}
A_{11} &= N_0 \\
A_{22} &= -A_{21} = \frac{\lambda_1}{\lambda_1 - \lambda_2} N_0 \\
A_{33} &= -A_{32} - A_{31} = \frac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} N_0 \\
&\cdot \\
&\cdot \\
&\cdot \\
&\text{etc.}
\end{aligned}
\qquad (10)
$$

Hence, it follows that the amount of any species k at time τ is simply

$$N_k(\tau) = N_0 [h_{k1} e^{-\lambda_1 \tau} + h_{k2} e^{-\lambda_2 \tau} + h_{k3} e^{-\lambda_3 \tau}$$

$$+ \ldots + h_{kk} e^{-\lambda_k \tau}]$$

where from Eqs. 8 and 10 and with a little rewriting the coefficients h_{ki} can be written in the following systematic way:

$$h_{k1} = \frac{\lambda_1}{\lambda_k - \lambda_1} \cdot \frac{\lambda_2}{\lambda_2 - \lambda_1} \cdot \frac{\lambda_3}{\lambda_3 - \lambda_1} \cdots \frac{\lambda_{k-1}}{\lambda_{k-1} - \lambda_1}$$

$$h_{k2} = \frac{\lambda_1}{\lambda_1 - \lambda_2} \cdot \frac{\lambda_2}{\lambda_k - \lambda_2} \cdot \frac{\lambda_3}{\lambda_3 - \lambda_2} \cdots \frac{\lambda_{k-1}}{\lambda_{k-1} - \lambda_2}$$

$$h_{k3} = \frac{\lambda_1}{\lambda_1 - \lambda_3} \cdot \frac{\lambda_2}{\lambda_2 - \lambda_3} \cdot \frac{\lambda_3}{\lambda_k - \lambda_3} \cdots \frac{\lambda_{k-1}}{\lambda_{k-1} - \lambda_3}$$

.
.
.

etc.

These relations are just the Bateman equations[4]. The above technique may also be applied to activation in a neutron field as is demonstrated in the following example.

B.   Example - simple model for (n,2n) activation.

Thermonuclear device performance is often diagnosed by measuring the ratio of second-order (n,2n) product to the first-order product for some loaded material. The processes are illustrated in Fig. 2, where the 3 nuclides may also be depleted by various neutron-induced destruction cross-sections, σ_{Di} (i=1,2,3). In a more realistic situation there are many more isotopes and isomers involved and also additional cross sections, such as (n,γ) which connect the species in the opposite direction to that shown in Fig. 2 for the (n,2n) reactions. Nevertheless, it has proved useful to develop a simple model which can be treated by the techniques developed here and which can be easily applied to understanding the general features of the diagnostics problems.

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In a nuclear explosion, the time scale is such that the second term in Eq. 3 is small relative to the first. For this simple model, we will drop the second term and, furthermore, we will assume that the neutron flux spectrum does not change its energy distribution in time. The consequences of this latter assumption will be examined later. It is equivalent mathematically to assuming that $\underset{\sim}{g}$ is independent of energy, solving Eq. 3 under that assumption, and then replacing $\underset{\sim}{g} \Phi (t)$ in the solution[5] by

$$< \underset{\sim}{g} \phi > \; = \int_{0}^{\infty} \underset{\sim}{g}(E)\Phi(t,E)dE$$

where $\Phi(t)$ and $\Phi(t,E)$ have been defined earlier. Hence, we write

$$\frac{d\vec{N}}{d\tau} = \underset{\sim}{g} \int_{0}^{\infty} \varphi(\tau,E)dE \cdot \vec{N} = \underset{\sim}{g}\varphi(\tau) \cdot \vec{N} \qquad (11)$$

and, recognizing that $\varphi(\tau)d\tau = d\Phi$, where

$$\Phi(t) = \int_{0}^{t} \varphi(\tau)d\tau$$

as defined previously, Eq. 11 becomes

$$\frac{d\vec{N}}{d\Phi} = \underset{\sim}{g} \cdot \vec{N} \qquad (12)$$

For the case illustrated in Fig. 2, the matrix $\underset{\sim}{\sigma}$ is

$$\underset{\sim}{\sigma} = \begin{pmatrix} \sigma_{11} & 0 & 0 \\ \sigma_{21} & \sigma_{22} & 0 \\ 0 & \sigma_{32} & \sigma_{33} \end{pmatrix}$$

where $\sigma_{11} = -\sigma_{D1} - \sigma_{21}$, with σ_{21} equal to the (n,2n) cross section connecting species 1 and 2. We assume that only species 1 is present at $\tau=0$. Applying the techniques of the last example, to which this is very similar, the results are:

$$N_1 = N_0 e^{<\sigma_{11}\phi>}$$

$$N_2 = \frac{N_0 <\sigma_{21}\phi>}{<\sigma_{11}\phi> - <\sigma_{22}\phi>} \left[e^{<\sigma_{11}\phi>} - e^{<\sigma_{22}\phi>} \right]$$

$$N_3 = \frac{N_0 <\sigma_{21}\phi> <\sigma_{32}\phi>}{<\sigma_{11}\phi> - <\sigma_{22}\phi>} \left[\frac{e^{<\sigma_{11}\phi>} - e^{<\sigma_{33}\phi>}}{<\sigma_{11}\phi> - <\sigma_{33}\phi>} - \frac{e^{<\sigma_{22}\phi>} - e^{<\sigma_{33}\phi>}}{<\sigma_{22}\phi> - <\sigma_{33}\phi>} \right]$$ (12')

where N_0 is the amount of loaded material i=1 at $\tau=0$.

2. Expansion Methods

The matrix method of the preceding section is easily adapted to a computer or can be used to generate analytic solutions for a few simple cases such as the examples illustrated above. It is particularly useful for cases where the fluence is great enough or the time long enough so that many multiple steps are involved.

The technique tends to mask the underlying structure of the problem and a development based on an expansion in, say, powers of the total fluence may be more transparent, although it may have convergence problems for very large fluences. A series solution for Eq. 3 is

$$\vec{N}(t) = [\underset{\sim}{1} + \int_0^t \underset{\sim}{D}(\tau)d\tau + \int_0^t \underset{\sim}{D}(\tau) \cdot \int_0^\tau \underset{\sim}{D}(\xi)d\xi d\tau +$$

$$\int_0^t \underset{\sim}{D}(\tau) \cdot \int_0^\tau \underset{\sim}{D}(\xi) \cdot \int_0^\xi \underset{\sim}{D}(\eta)d\eta d\xi d\tau + \cdots] \cdot \vec{N}_0,$$

$$\text{where } \underset{\sim}{D}(\tau) = \int_0^\infty \underset{\sim}{\sigma}(E)\varphi(\tau,E)dE + \underset{\sim}{a} \cdot \underset{\sim}{\lambda} \quad .$$

This so-called time-ordered series[6] is exact, as can be seen by direct substitution into the differential equation. For the case where the second term, $\underset{\sim}{a} \cdot \underset{\sim}{\lambda}$, of Eq. 3 is small compared to the first, the above solution reduces to

$$\vec{N}(t) = \left[\underset{\sim}{1} + \int_0^\infty \underset{\sim}{\sigma}(E) \int_0^t \varphi(\tau,E)d\tau dE + \right.$$

$$\int_0^\infty \int_0^\infty \underset{\sim}{\sigma}(E) \cdot \underset{\sim}{\sigma}(E') \int_0^t \varphi(\tau,E) \int_0^\tau \varphi(\xi,E')d\xi d\tau dE' dE$$

$$\left. + \cdots \right] \cdot \vec{N}_0 \quad . \tag{13}$$

Equation 13 is not very useful in this form; however, we note
that the second term is just $<g\ \Phi>$ as defined for the simple
model in Part 1.B above. This term arose because of the
assumption that the flux spectral shape was independent of time.
It followed that the final result depended only on the total
fluence spectrum to time t, $\Phi(t,E)$ and not on its time history.
For the exact solution, Eq. 13, we see that this statement is true
only to first order. The second order term correlates energies E
and E' as a function of time and depends not just on the total
fluence spectrum, but on the flux time history. Because, however,
this correlation first enters in second order, it is less
important for some reactions than for others. The importance of
these correlations will be examined later.

If, again, we assume that the flux spectral shape is
independent of time, i.e., that $\varphi(\tau,E) = g(\tau)f(E)$, then
Eq. 13 reduces to

$$\vec{N} = e^{<g\ \Phi>} \cdot \vec{N}_0 = [1 + <g\ \Phi> + \frac{1}{2!}<g\ \Phi>^2 + \frac{1}{3!}<g\ \Phi>^3$$
$$+ \cdots \quad] \cdot \vec{N}_0$$

$$(14)$$

where the matrix exponential is defined by the series expansion in
brackets. The expression $<g\ \Phi>^3$ stands for $<g\ \Phi> \cdot <g\ \Phi> \cdot <g\ \Phi>$,
etc. and $<g\ \Phi>$ is as previously defined.

The various activation processes taking place in a strong
neutron flux can be understood on the basis of the various terms
in Eq. 14. This is illustrated in Fig. 3, in which we consider
the irradiation of ^{90}Zr. The unit matrix in Eq. 14 connects

only to loaded material, so that the leading term for production of ^{89}Zr (by the (n,2n) reaction) is the first-order term $\langle \underset{\sim}{\sigma} \Phi \rangle$. Production of ^{88}Zr, on the other hand, proceeds through two successive (n,2n) reactions as shown in the top part of Fig. 3 and depends therefore on $\langle \underset{\sim}{\sigma} \Phi \rangle^2$ in leading order. Similarly, depletion of ^{89}Zr by neutron-induced reactions (shown schematically in the lower part of Fig. 3) is also second order, $\langle \underset{\sim}{\sigma} \Phi \rangle^2$. In order to produce ^{86}Zr from ^{90}Zr by successive (n,2n) reactions the leading term is $\langle \underset{\sim}{\sigma} \Phi \rangle^4$. Consequently, because of the strong dependence of such high-order products on neutron fluence, they are candidates for diagnosing regions of very high fluence. However, the actual amount of fourth-order product depends not only on the fluence as indicated above, but on many different combinations of cross sections through the matrix product $\underset{\sim}{\sigma}^4$. In addition, recalling that Eq. 14 is only an approximation, the result will also depend on the flux time-history. We need, therefore, to examine both the sensitivity to cross-section values and to the evolution of the fluence spectrum with time.

3.   Expansion of Exact Solution

The expansion shown in Eq. 13 is exact, but it is not in a very useful form. Equation 14, on the other hand, is not very exact, but is quite useful for making simple interpretations (see Sec. IV). We wish now to develop the connection between these two equations. Referring to Eq. 3, again without the second term, namely

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$$\frac{d\vec{N}}{d\tau} \approx \int_0^\infty \underset{\sim}{\sigma}(E)\ \varphi(\tau,E)dE \cdot \vec{N} \qquad (15)$$

we define $\underset{\sim}{\sigma}(\tau)$ according to

$$\underset{\sim}{\sigma}(\tau) = \frac{\displaystyle\int_0^\infty \underset{\sim}{\sigma}(E)\ \varphi(\tau,E)dE}{\displaystyle\int_0^\infty \varphi(\tau,E)dE} \qquad (16)$$

This "energy-averaged cross section" should have only a weak time-dependence; it is, in fact, independent of time if $\varphi(\tau,E)$ is factorable. We may write Eq. 15 then as

$$d\vec{N} = \underset{\sim}{\sigma}(\tau)\ \cdot\ \vec{N}(\tau)\ \varphi(\tau)d\tau \qquad (17)$$

where

$$\varphi(\tau) = \int_0^\infty \varphi(\tau,E)dE$$

as previously defined. Recognizing that $d\Phi(\tau) = \varphi(\tau)d\tau$, Eq. 17 becomes

$$\frac{d\vec{N}}{d\Phi} = \underset{\sim}{\sigma}(\Phi)\ \cdot\ \vec{N}(\Phi) \qquad (18)$$

where Φ is the total fluence and $\underset{\sim}{\sigma}(\Phi)$ is $\underset{\sim}{\sigma}(\tau)$ evaluated at a time τ for which the total fluence to that time is Φ. Because of the structure of Eq. 16 the dependence of $\underset{\sim}{\sigma}(\Phi)$ on Φ will be weak and a series expansion in powers of Φ will be reasonable, namely

$$\underset{\sim}{\alpha}(\phi) = \sum_k \underset{\sim}{\alpha}_k \phi^k \quad , \qquad \underset{\sim}{\alpha}_0 = \underset{\sim}{\alpha}(0), \ \underset{\sim}{\alpha}_1 = \frac{d\underset{\sim}{\alpha}}{d\phi}\bigg|_0 \quad , \ \text{etc.}$$

We assume a solution for $\vec{N}$,

$$\vec{N}(\phi) = \sum_n \vec{a}_n \phi^n \quad , \quad \vec{N}(0) = \vec{N}_0 = \vec{a}_0$$

Substituting these series into Eq. 18 yields a recurrence relation for the expansion vectors $\vec{a}_n$,

$$(n+1)\vec{a}_{n+1} = \sum_{k=0}^{n} \underset{\sim}{\alpha}_{n-k} \cdot \vec{a}_k \tag{19}$$

Using Eq. 19 to evaluate the first few terms in $\vec{N}(\phi)$ one may write the solution in such a way as to compare Eqs. 13 and 14.

$$\vec{N}(\phi) = e^{<\underset{\sim}{\alpha} \ \phi>} \cdot \vec{N}_0 + \frac{\phi^3}{12} \ [\underset{\sim}{\alpha}_1, \underset{\sim}{\alpha}_0] \cdot \vec{N}_0$$

$$+ \frac{\phi^4}{24} \left\{ 2 \ [\underset{\sim}{\alpha}_2, \underset{\sim}{\alpha}_0] + [\underset{\sim}{\alpha}_1, \underset{\sim}{\alpha}_0]^2 \right\} \cdot \vec{N}_0 + \cdots \tag{20}$$

where $\vec{N}$ represents the exact solution, Eq. 13, and the leading term on the right hand side is just the approximate solution, Eq. 14. The square brackets in Eq. 20 are commutators, i.e., $[\underset{\sim}{A}, \underset{\sim}{B}] = \underset{\sim}{A} \cdot \underset{\sim}{B} - \underset{\sim}{B} \cdot \underset{\sim}{A}$, and are a measure of the importance of the flux time history for each order. For example, suppose that because of the cross sections involved, the elements of the commutator $[\underset{\sim}{\alpha}_1, \underset{\sim}{\alpha}_0]$ which contribute to a given product N_j are negligible. Then, to second order, the production of

N_j will depend only on the total time-integrated fluence and not its history. This result may not obtain for some other species N_i because of differences in the cross sections. This would mean, then, that the product N_j is less sensitive to the details of the neutron production and therefore the particular computational modeling that went into generating the neutron flux, whereas N_i may be very sensitive.

In the above development the key factor was changing from time as the independent variable to fluence Φ. In fact, one may follow exactly the same procedure and change the independent variable to any special fluence; e.g. the "14-MeV fluence" may be particularly useful for fusion reactors or thermonuclear weapons. As a result certain products, e.g. those with (n,2n) thresholds ≤ 14 MeV, correlate most strongly with the total 14-MeV fluence and are very insensitive to the time-development of other parts of the spectrum. Consequently, the above formalism may be used to look for and uncover important correlations -- for some applications one may desire sensitivity to details, for others lack of sensitivity may be important. In the companion paper[2], this approach is applied to some specific thermonuclear diagnostic problems. Here we will continue to develop general techniques.

4. Spatial Dependence

The equations developed above apply to a zone within which the neutron fluence was considered to be spatially uniform. We write for the production vector in this zone $d\vec{N} = \underline{\underline{D}} \cdot d\vec{N}_0$, where $d\vec{N}_0$ represents the initial population in the zone and $\underline{\underline{D}}$

represents a matrix operator such as the square brackets in
Eqs. 13 or 14. The fluxes appearing in the various expressions
for $\underset{\sim}{D}$ are just for the given zone. Consider now a region composed
of many zones in which the loaded material is distributed
uniformly. Let $\overline{N}_0$ be the total initial population in the loaded
region and M the total mass. It follows that

$$d\overline{N}_0 = \overline{N}_0 \frac{dM}{M}$$

where dM is he mass of a zone. (We construct zones and regions
so that M and dM are constant in time.) Consequently,

$$\overrightarrow{N} = \int\limits_{\text{region}} d\overrightarrow{N} = \left[\int\limits_{\text{region}} \underset{\sim}{D} \cdot \frac{dM}{M} \right] \cdot \overrightarrow{N}_0 \tag{21}$$

The leading term in the bracket in Eq. 21 is just the unit matrix
and the second term is

$$\int\limits_{\text{region}} <\underset{\sim}{\sigma}\ \Phi> \frac{dM}{M} = <\underset{\sim}{\sigma}\ \overline{\Phi}> \quad \text{, where}$$

$\overline{\Phi}$ is the average fluence spectrum in the region

$$\overline{\Phi} = \frac{1}{M} \int\limits_{\text{region}} \Phi(t,E)dM$$

Recalling that the first-order term is the same in either Eq. 13
or 14 (see Eq. 20) it is important to notice that to first-order
in Φ, one-step processes are independent of the flux time-history
and depend only on the zonal average of the fluence spectrum.
Using approximate Eq. 14 for the next term in $\underset{\sim}{D}$, we have

$$\frac{1}{2} \int_{region} <\underset{\sim}{\sigma} \, \Phi>^2 \, \frac{dM}{M} = \frac{1}{2} \int_0^{\infty} \int_0^{\infty} \underset{\sim}{g}(E) \cdot \underset{\sim}{g}(E') \, [\int_{region} \Phi(t,E) \, \Phi(t,E') \, \frac{dM}{M}] \, dEdE'$$

where the quantity in brackets is again a correlation function correlating energies E and E' as a function of spatial coordinate, i.e., zone. If, however, the fluence spectral shape is spatially uniform, then the correlation is destroyed and the second order term reduces to $\frac{1}{2} <\underset{\sim}{\sigma} \, \Phi_{RMS}>^2$, where

$$\Phi_{RMS} = [\frac{1}{M} \int_{region} \Phi^2(t,E)dM]^{1/2} \quad .$$

In general, however, one finds that spectral shape changes across a region may be substantial so that products of 2-step processes, e.g., ^{88}Zr, may be affected by spectral correlations induced not only by the flux time history, but also as a result of spatial non-uniformities. Such is not the case for the first order products, such as ^{89}Zr. By appropriate loading of materials into spatially restricted regions, the sensitivity will be limited to gross features of the fluence distributions, $\bar{\Phi}$, Φ_{RMS}, etc.

IV. APPLICATION TO RADIOCHEMICAL DETECTORS: SENSITIVITY TO CROSS SECTIONS.

As mentioned earlier, certain materials, often those with high (n,2n) thresholds, are loaded into thermonuclear devices and their activation products which result after detonation are recovered and

used to diagnose device performance. Typically, from the debris of an underground nuclear test one measures the ratio of one residual activity to another. The measured ratios are compared to calculations performed with activation codes, such as WATUSI[7], which employ neutron fluences generated from explosion codes. In order to make the activation calculations as realistic as possible, the set K of isotopes and isomers employed is often very large so that the number of cross sections required may approach a hundred or more. Most of these cross sections are for the unstable products created in the neutron flux and therefore are not measured directly. Instead, it is necessary to rely on calculations based on the appropriate nuclear models.[8] Depending on energy, type of reaction and nuclear structure information the reliability of these calculations can vary enormously. The interpretation of a certain measured detector ratio may be suspect unless the sensitivity of the result to cross section uncertanties is known. Some cross sections may be more important +han others and it is important to establish a framework which allows attention to be focused on the most sensitive reactions. The formalism developed above allows this to be done.

Let $R^{k\ell} \equiv N_k/N_\ell$ be the desired isotope ratio at time t (or after fluence Φ) -- the ratio of the second order (n,2n) product ^{88}Zr to the first order product, ^{89}Zr, for loaded natural Zr (51.5% ^{90}Zr, 11.2% ^{91}Zr, 17.1% ^{92}Zr, 17.4% ^{94}Zr, and 2.8% ^{96}Zr) is a realistic example. The sensitivity of this ratio to any particular cross section σ_{ij}, may be defined as the fractional change in $R^{k\ell}$ resulting from a given fractional change in σ_{ij}, or

$$\beta\,_{ij}^{k\ell} = \frac{\sigma_{ij}}{R^{k\ell}}\,\frac{\partial R^{k\ell}}{\partial \sigma_{ij}} = \beta_{ij}^{k} - \beta_{ij}^{\ell} \quad , \text{ where}$$

$$\beta_{ij}^{\ell} = \frac{\sigma_{ij}}{N_{\ell}}\,\frac{\partial N_{\ell}}{\partial \sigma_{ij}} \tag{22}$$

Because we are interested in general guidelines rather than high precision (which may be accomplished by a computer) we will assume that spatial gradients and time history are unimportant for the purposes of cross section sensitivity calculations. Consequently, for a given loaded isotope N_{mo}, it follows that

$$N_{\ell} = [\delta_{\ell m} + \sigma_{\ell m}\,\Phi + \frac{1}{2}\,\sigma_{\ell m}^{2}\,\Phi^{2} + \dots]\,N_{mo} \tag{23}$$

where Eq. 23 follows from Eq. 14, Φ is the total fluence, $\delta_{\ell m}$ is the Kronecker δ-function, and $\sigma_{\ell m}$ is the cross section for the reaction $m \rightarrow \ell$ averaged over the assumed constant spectral shape of the neutron flux. The quantity $\sigma_{\ell m}^{2}$ means matrix multiplication $\sum\limits_{k} \sigma_{\ell k}\,\sigma_{km}$, etc. From Eq. 23 it follows that

$$\frac{\partial N_{\ell}}{\partial \sigma_{ij}} = [\xi_{\ell m}^{ij}\,\Phi + \frac{1}{2}\,\zeta_{\ell m}^{ij}\,\Phi^{2} + \frac{1}{6}\,n_{\ell m}^{ij}\,\Phi^{3} + \dots]\,N_{mo} \tag{24}$$

where
$$\xi_{\ell m}^{ij} = \frac{\partial \sigma_{\ell m}}{\partial \sigma_{ij}}$$

$$\zeta_{\ell m}^{ij} = \frac{\partial \sigma_{\ell m}^{2}}{\partial \sigma_{ij}} = \sum\limits_{k} [\sigma_{\ell k}\,\xi_{km}^{ij} + \xi_{\ell k}^{ij}\,\sigma_{km}]$$

and
$$n_{\ell m}^{ij} = \frac{\partial \sigma_{\ell m}^{3}}{\partial \sigma_{ij}} = \sum\limits_{k} [\sigma_{\ell k}\,\zeta_{km}^{ij} + \zeta_{\ell k}^{ij}\,\sigma_{km}] \, , \quad \text{etc.}$$

Using these relations and the definition, Eq. 2, for the diagonal elements σ_{ij}, it follows that

$$\xi^{ij}_{\ell m} = -\delta_{\ell m}\delta_{j\ell} + (1-\delta_{\ell m})\delta_{i\ell}\delta_{jm} \tag{25}$$

$$\zeta^{ij}_{\ell m} = -\sigma_{jm}\delta_{i\ell} + \sigma_{\ell i}\delta_{jm} - \sigma_{\ell m}[\delta_{jm}(1+\delta_{im}) + \delta_{j\ell}(1+\delta_{i\ell})]$$

$$\eta^{ij}_{\ell m} = \sigma^2_{jm}\delta_{i\ell} + \sigma^2_{\ell i}\delta_{jm} - \sigma^2_{\ell m}[\delta_{jm}(1+\delta_{im})+\delta_{j\ell}(1+\delta_{i\ell})]$$

$$+ \sigma_{jm}\lceil\sigma_{\ell i} - \sigma_{\ell j}(1+\delta_{ij})\rceil$$

From the relations in Eq. 25 it is straightforward to calculate the sensitivities β^{ℓ}_{ij} for any assumed set of cross sections and total fluence Φ. A particularly useful example is that for threshold (n,2n) detectors where one loaded isotope (or a natural material with one predominant isotope) produces first, second and higher order products by multiple (n,2n) reactions, but (n,3n) reactions are not allowed energetically. Figure 4 illustrates this example where m labels the loaded isotope, m + 1 the first-order product and m + 2 the second. In addition to the (n,2n) reactions shown, there are (n,γ) reactions coupling in the opposite direction and there may be several isomeric states (i,j,k..) coupled by (n,2n), (n,γ), (n,n') reactions to the states shown and to each other. The processes for which sensitivities are calculated are shown in Fig. 5 where i labels one of the isomeric states, the arrow represents the particular reaction whose cross section is being examined and the heavy horizontal line is the state being measured. Simple formulas for the processes shown in Fig. 5 are given in Table I. These formulas are

given only to leading order (or first order in Φ) and apply to any case which satisfies the above conditions for a threshold (n,2n) detector. They have been compared against explosion code calculations and have been found to be very close to the computer calculations[2] and therefore may serve a useful guide. They apply to as large a cross section set as desired, and for certain restricted sets many of the formulas reduce considerably. (For the formulas given, i may be set equal to D and σ_{Dj} will represent (n, dest) on j, σ_{jD} is defined to be zero. Furthermore, it is to be stressed that σ^2_{ij} stands for $\sum_k \sigma_{ik}\sigma_{kj}$.)

Several points are evident from examining Table I. For example, the leading term in the sensitivity of the first order product to the primary (n,2n) cross section is unity, independent of fluence. A first order correction term tends to reduce the sensitivity at high fluences, nevertheless this cross section is the most sensitive. At low fluences a 10% change in the primary (n,2n) cross section means a 10% change in the results. Consequently, this cross section requires the best accuracy of any in the set if the final results are to be reliable. Fortunately, because it is a cross section on a stable isotope, it can be measured. Other sensitivities which are independent of fluence in leading order are $\beta^{m+2}_{m+2, i}$ and β^{m+2}_{im} for the population of the second-order product for any 2-step process connecting the loaded isotope with the detected second-order products. These processes involve cross sections on unstable nuclides, e.g., (n,2n) on the first order product, or isomers which unlike the primary (n,2n) cross section are difficult or often impossible to measure. Consequently, uncertainties in the

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modeling of these cross sections may put severe limits on the accuracy of the N_2/N_1 ratio which persist even at very low fluences. (The first-order term in Φ for these sensitivities is straightforward to calculate, but for simplicity is not included in Table I.)

All other sensitivities illustrated in Fig. 5 are proportional to the total fluence in leading order. Consequently, they become more important to overall accuracy as fluence is increased. Nevertheless, there are some general observations which can be made. For example, the sensitivities of either first-order or second-order products to any reaction depleting those products $\beta_{i,m+1}^{m+1}$, $\beta_{i,m+2}^{m+2}$ are simply proportional to the cross section for the particular depletion reaction regardless of the complexity of the cross section set. Consequently, the importance of neutron capture on the second-order product, for example, may be quickly estimated. The formulas given in Table I allow the sign of the the sensitivity to be determined, regardless of cross section, in all but a few cases. For correlated errors in the calculated cross sections, knowledge of the algebraic sign may pinpoint cancellations or enhancements which can affect the overall error. Lastly, some reactions only affect a given product in second-or higher powers of the fluence, i.e. $\beta = 0$ in Table I. Cross sections for such reactions will be considerably less important and can probably be ignored.

The formulas in Table I are approximate, but have been found to be reasonably accurate even at high fluence when compared to exact computer activation calculations using neutron fluence files from explosion code calculations for actual devices. Specific comparison

will be given in the companion paper.[2] These formulas, therefore, may be used to make quick estimates or to serve as simple guides to direct more complicated computer calculations.

V.   CONCLUSION

This paper has developed the general formalism for the multiple-step activation and decay of nuclides in a neutron flux. Exact, general results have been derived in terms of a series expansion and various limits have been studied. Techniques for handling spatial gradients and flux time-history have been developed. Specialized solutions in terms of average quantities have been presented.

These general techniques were applied to a simple example of determining the sensitivity of the production of first or second order (n,2n) products to various of the many cross sections. Such sensitivity results have direct application to radiochemical thermonuclear detectors and to determining the relative importance and required accuracy of the many different cross sections.

In a companion classified paper[2], many of the techniques and results developed here will be applied to specific thermonuclear weapon problems and direct comparisons to computer-generated results will be made.

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3. In general, the matrix $\underline{B}$ will be real, but not necessarily symmetric or
    orthogonal. Consequently, whether any given case can be diagonalized
    requires analysis of the secular equation $\det\left|\underline{B} - \gamma\underline{1}\right| = 0$, the
    solutions of which may be multiple and complex.

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5. This may be seen most easily by writing $\varphi(\tau,E)$ as a product $f(\tau)g(E)$
    and substituting this into Eq. 3. The equation which results will have
    the same solutions as Eq. 12, but with $\underline{g}\phi$ replaced with its
    energy-average, $<\underline{g}\,\phi>$.

6. The equation for $\vec{N}(t)$ may also be written $\vec{N}(t) = \mathrm{Texp}\left\{\int_0^t \underline{D}(\tau)d\tau\right\} \cdot \vec{N}_0$,
    where T is the time-ordering operator. The concept of time-ordering is
    discussed by M. L. Goldberger and K. M. Watson, Collision Theory,
    John Wiley and Sons, Inc., New York, N.Y. (1965), p. 48

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8. C. H. Poppe, "Nuclear Modeling and Its Application to Nuclear Weapon
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    1979) p. 7.

TABLE I. Sensitivity for processes illustrated in Figure 5.

Sensitivity	Comments
$\beta^{m}_{im} = -\sigma_{im}\,\Phi$, all $i \neq m$	Depletion of loaded isotope.
$\beta^{m+1}_{m+1,m} = [1 - \frac{1}{2}\left(\sigma_{m+1,m} + \sigma^{-1}_{m+1,m} \sum\limits_{\substack{i\neq m \\ i\neq m+1}} \sigma_{m+1,i}\,\sigma_{im}\right)\Phi]$	Primary production cross section $(n,2n)$
$\beta^{m+1}_{i,m+1} = -\frac{1}{2}\sigma_{i,m+1}\,\Phi$, all $i \neq m+1$	Any reaction that depletes first-order product.
$\beta^{m+1}_{m+1,i} = \frac{1}{2}\frac{\sigma_{m+1,i}\,\sigma_{im}}{\sigma_{m+1,m}}\,\Phi$, all $i \neq m,\ m+1$	Any reaction other than primary production $(n,2n)$ populating *first-order product*
$\beta^{m+1}_{im} = \frac{1}{2}\frac{\sigma_{im}(\sigma_{m+1,i}-\sigma_{m+1,m})}{\sigma_{m+1,m}}\,\Phi$, all $i \neq m, m+1$	Any reaction other than primary production $(n,2n)$ depleting loaded isotope.

Table I - Continued

$\beta_{mi}^{m+1} = 0$, all $i \neq m, m+1$

Any reaction populating loaded other than (n, γ) from first-order product.

$\beta_{ij}^{m+1} = 0$, all $i \neq m, m+1, j \neq m, m+1$

Any reaction connecting neither loaded isotope nor first-order product.

$\beta_{m+2,i}^{m+2} = \dfrac{\sigma_{m+2,i} \, \sigma_{im}}{\sigma_{m+2,m}^{2}}$, all $i \neq m, m+2$

Production of second order product by any 2-step process from loaded isotope.

$\beta_{im}^{m+2} = \dfrac{\sigma_{m+2,i} \, \sigma_{im}}{\sigma_{m+2,m}^{2}}$, all $i \neq m, m+2$

Reactions depleting loaded isotope and connected to second-order product.

$\beta_{i,m+2}^{m+2} = -\dfrac{1}{3} \sigma_{i,m+2} \, \Phi$, all $i \neq m+2$

Any reaction depleting second-order product.

Table I - Continued

$$\beta_{m+2,i}^{m+2} = \frac{\sigma_{m+2,i}\,\sigma_{i,m+1}}{3\,\sigma_{m+2,m+1}}\,\Phi \ , \ \text{all } i \neq m+1, m+2$$

Any reaction which is part of a 3-step process populating second-order product (i.e. if $\sigma_{im} = 0$).

$$\beta_{i,m+1}^{m+2} = \frac{\sigma_{i,m+1}\,\sigma_{m+1,m}\,(\sigma_{m+2,i} - \sigma_{m+2,\,m+1})}{3\sigma_{m+2,m}^{2}}\,\Phi \ ,$$

$$\text{all } i \neq m+1, m+2$$

Any reaction depleting first-order product not connected to second-order.

$$\beta_{m+1,i}^{m+2} = \frac{\sigma_{m+1,i}\,\sigma_{im}\,(\sigma_{m+2,m+1} - \sigma_{m+2,i})\,\phi}{3\sigma_{m+2,m}^{2}}$$

$$\text{all } i \neq m, m+1, m+2$$

Any reaction populating first order, but not from loaded isotope or second order.

$$\beta_{mi}^{m+2} = \frac{\sigma_{im}(\sigma_{m+1,i} - \sigma_{m+1,m})\,\Phi}{3\sigma_{m+1,m}} \ , \ \begin{array}{l} \text{all } i \neq m , \\ m+1, m+2 \end{array}$$

Any reaction which is part of a 3-step process to second-order which depletes loaded isotope (i.e. $\sigma_{m+2,\,i} = 0$)

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Table I - Continued

$$\beta_{mi}^{m+2} = -\frac{\sigma_{mi}\,\sigma_{im}\,\sigma_{m+2,i}}{3\sigma_{m+2,m}^2}\,\Phi\ ,\quad \text{all } i \neq m, m+2$$

Any reaction populating loaded isotope.

Figure Captions

Figure 1   Simple radioactive decay chain with parent N_1. For this example
            $\lambda_{ij} = \lambda_i$ the total decay constant of nuclide N_i .

Figure 2   Simple (n,2n) activation chain which ignores decay and (n,γ)
            reactions back down the chain. Any process other than (n,2n)
            which depletes a given nuclide is included in "Destruction."

Figure 3   Examples of 2-step processes: ^{88}Zr production (top) and ^{89}Zr
            destruction (bottom).

Figure 4   Realistic activation chain where loaded isotope m connects to
            first-order (n,2n) product m+1, which connects to second-order
            product m+2, etc. Feedback by (n,γ) is included as well as
            (n,n$'$) to isomeric states, i,j,k etc. Coupling through isomers
            is allowed as well as neutron-induced processes, (n, dest), which
            lead to nuclides not included in the set. Actual problems can be
            considerably more complicated than this.

Figure 5   Schematic diagrams illustrating the meaning of various
            cross-section sensitivity functions, β_{ij}^{k}. The horizontal
            lines labled m,m+1 and m+2 stand for the loaded isotope, first-
            order (n,2n) product and second-order product, respectively--i
            stands for any other state or isomer (for i=0, the corresponding
            cross section, $\underline{e}.\underline{g}.$ σ_{Dm}, would represent (n, dest.) processes.

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The heavy horizontal line represents the nuclide k being measured and the arrow stands for the cross section σ_{ij} with respect to which the sensitivity β^k_{ij} is determined.
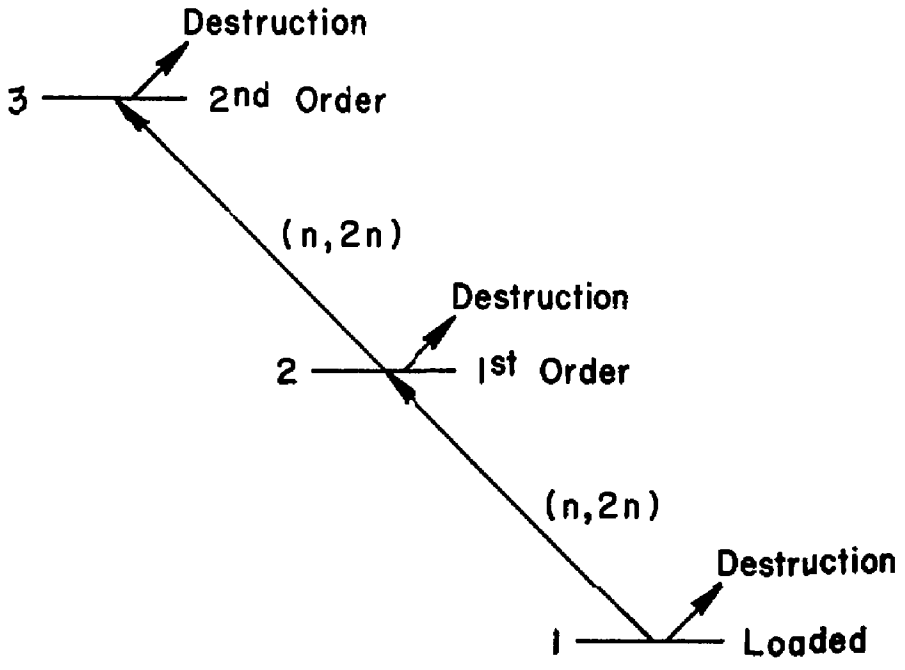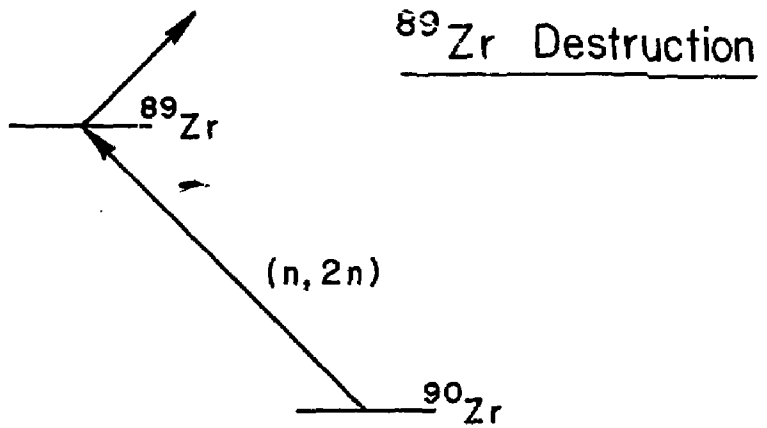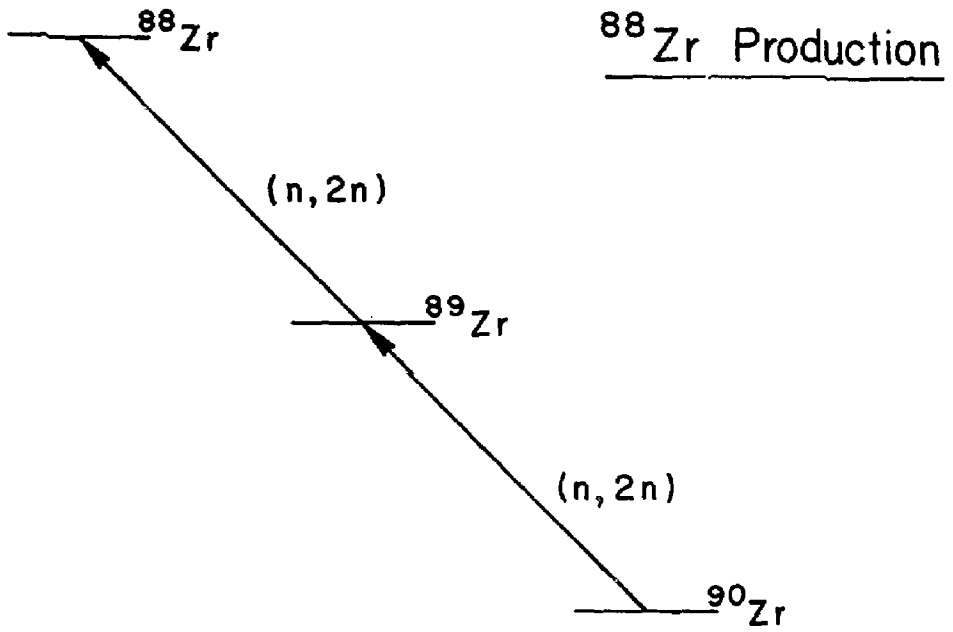
Figure 1

Destruction

3 ——————— 2nd Order

(n,2n)

Destruction

2 ——————— 1st Order

(n,2n)

Destruction

1 ——————— Loaded

Figure 2

^{88}Zr Production

^{88}Zr

(n, 2n)

^{89}Zr

(n, 2n)

^{90}Zr

^{89}Zr Destruction

^{89}Zr

(n, 2n)

^{90}Zr
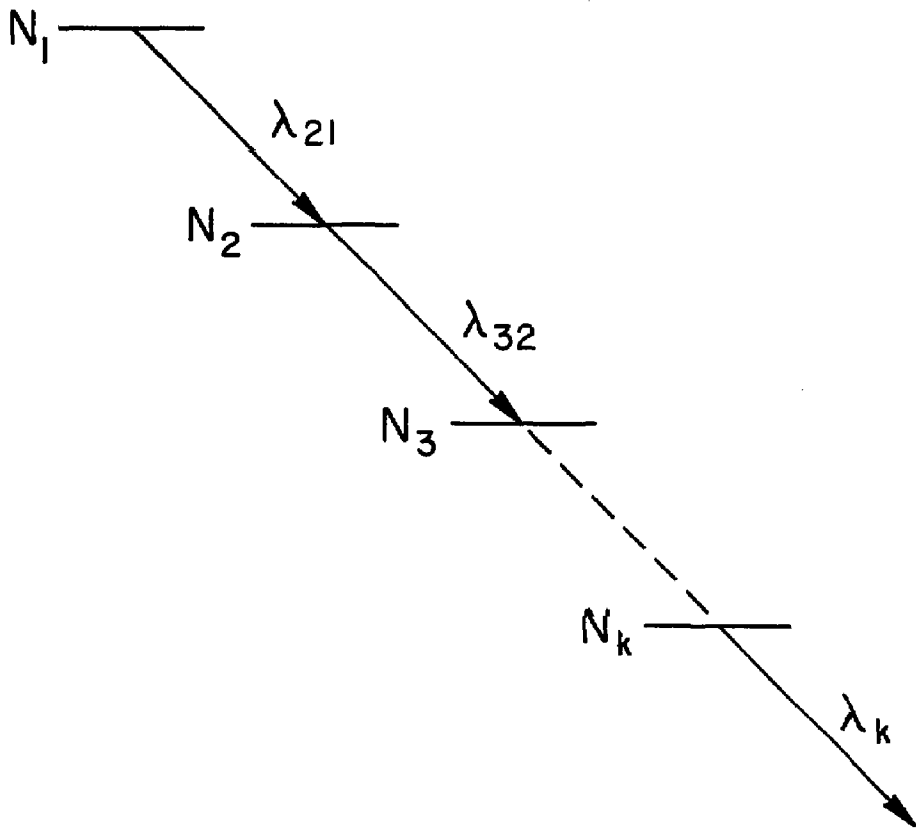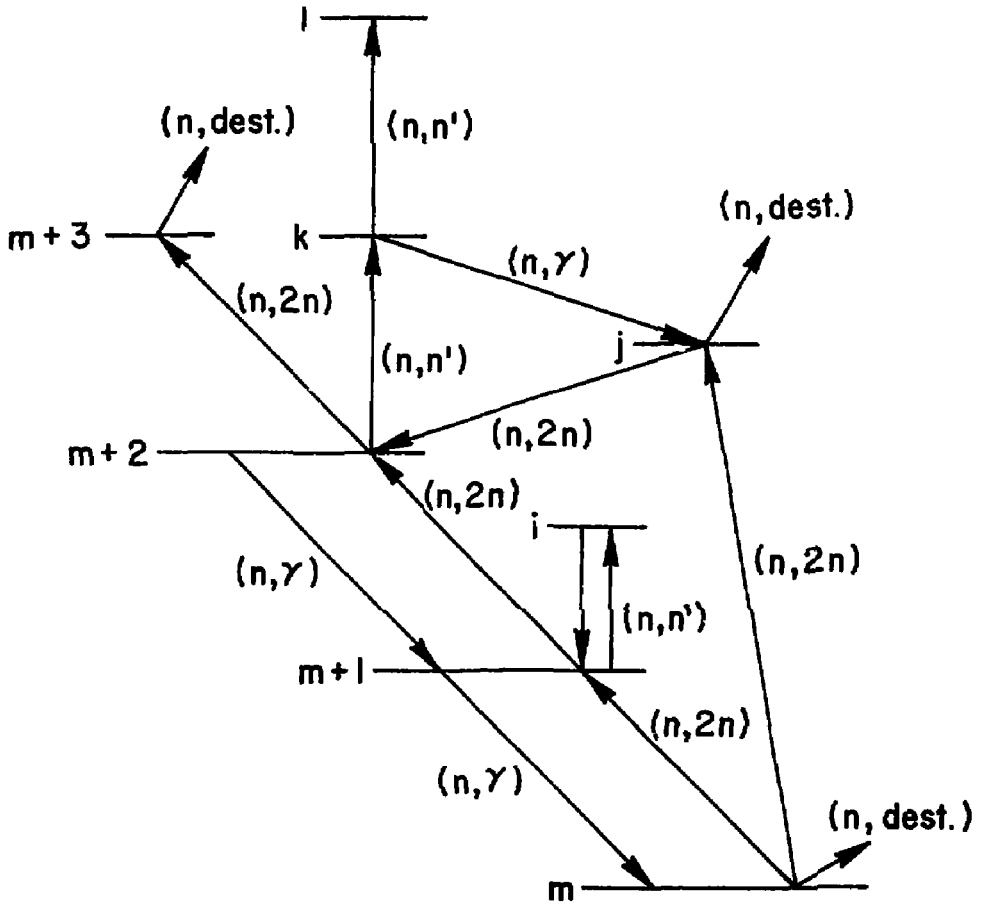
Figure 3

Figure 4

Figure 5

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