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METHODS OF CALCULATING YIELDS IN CHAINS OF SUCCESSIVE NUCLEAR TRANSFORMATIONS

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ABSTRACT

The mathematical problem of calculating the yield of an isotope produced in a chain of first order transformation processes is discussed.

Several new approximation formulas are derived, as well as the well-known general exact solution.

A sample calculation of the pile yield of isotopes in a chain of radiative neutron capture processes is carried out, illustrating a method of tabulation designed to minimize numerical errors.

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INTRODUCTION

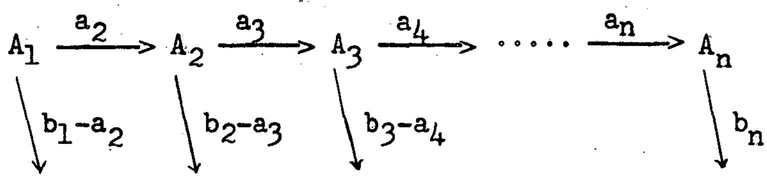
The mathematical problem of the variation with time of quantities involved in chains of first order transformations (as radioactive growth and decay and nuclear transformations effected by bombardment of matter by neutrons, charged particles, or gamma rays) has been treated by several authors.[1-5]

In connection with some of the experimental work at the University of California Radiation Laboratory it has been necessary to make extensive use of this type of calculation. Some new series expansions have been developed for use in certain cases. It was felt that it would be worth while to set down our general methods of setting up such problems, to list labor-saving formulas and to illustrate a method of tabulation, designed to minimize the possibility of error.

Since the first order differential equations in any complex chain constitute a set of linear equations, we can immediately divide any complex chain with any distribution of initial quantities into sums of unbranched chains with only the parent initially present. The calculations can be carried through individually and the summation numerically performed as the last step.

PROBLEM OF THE SINGLE CHAIN WITH ONLY THE PARENT INITIALLY PRESENT

Consider a chain of first order transformations (as a series of n,γ reactions at constant neutron flux* or a radioactive decay chain):



where a_i is the constant coefficient governing the rate of production of isotope A_i from its parent A_{i-1}. If this production is by radioactive decay,

$$a_i = \lambda_i = 0.693/\text{half-life}$$

of A_{i-1} in seconds; and if the production is a bombardment particle-induced reaction, then $a_i = I\sigma_i$, where I is the flux of bombarding particles in particles per square centimeter per second, and σ_i is the capture cross section of A_{i-1} in square centimeters.

b_i is the constant coefficient governing the rate of destruction of isotope A_i by all processes. It is then the sum of all ${}_i\lambda_j$ for radioactive decay of A_i by all modes j plus the sum of all $I({}_i\sigma_k)$ for all particle-induced reactions k destroying A_i where σ_i is the

---

*If no radioactive decay processes are significant in the chain, the condition of constant neutron flux need not be imposed. The total integrated flux (nvt) is all that need be known. But if radioactive decay processes are significant, the flux variations give to the set of differential equations constants variable with time. If the variations can be treated as stepwise constant, the problem may be broken up and treated in separate segments. A suitable polynomial in time t might be fitted to the flux variation and the resulting differential equations in turn treated by the Laplace transformation method. We shall not, however, be concerned with any but the constant flux case here.

cross section of A_i in square centimeters for process k. That is,

$$b_i = \sum_j {}_i\lambda_j + I \sum_k {}_i\sigma_k \ .$$

The dimensions of the a_i and b_i are reciprocal time.

The differential equations relating the amounts y_i of species A_i present* are:

$$dy_1/dt = -b_1 y_1$$
$$dy_2/dt = a_2 y_1 - b_2 y_2$$
$$\cdots\cdots\cdots\cdots$$
$$dy_n/dt = a_n y_{n-1} - b_n y_n \ .$$

The following text will describe the derivations of the various equations used in the subject problem, beginning with the above relationships. The reader interested mainly in the practical application of the equations for solving a specific problem may turn directly to the illustrative example at the end of the paper.

With the initial conditions at t = 0,

$$y_1 = A$$
$$y_i = 0, \quad \text{for } i \neq 1$$

the first equation above is readily integrated to give the familiar exponential decay

$$y_1 = Ae^{-b_1 t} \ .$$

---

*The quantity y_i may be number of atoms, number of moles, or other convenient measure of number of atoms.

This expression can be substituted into the second equation and another integration performed to give the growth- and decay-type yield:

$$y_2 = Aa_2 \frac{e^{-b_1 t} - e^{-b_2 t}}{b_2 - b_1} .$$

This process could be done repeatedly to solve any such system, but it soon becomes cumbersome. It is thus much more convenient to apply the methods of operational calculus to such a system of differential equations. See Appendix I for the derivation.

For convenience and accuracy in the numerical calculations it seemed best to introduce dimensionless parameters of magnitudes near unity to replace the time, cross section, and flux variables. We let $q_i = ta_i$ and $C_i = tb_i$.

If ${}_tN_k$ is the number of atoms of the kth chain member at time t and ${}_0N_1$ is the number of atoms of the first member at time zero, we may write

$${}_tN_k = {}_0N_1 \cdot \prod_{2 \leq i \leq k} q_i \cdot \left[C_1, C_2, \ldots, C_n\right]$$

where $\left[C_1, C_2, \ldots, C_n\right]$ is a quantity designated as the "C-bracket."

A few formulas for evaluating C-brackets are listed below. The derivations in the appendices to this paper should suggest other means of dealing with special cases that may be encountered.

A sample of actual neutron pile transformation calculations is given to illustrate our method of tabulation. The results of this sample calculation are plotted on a log-log plot of yield versus integrated neutron flux.

USEFUL FORMULAS FOR THE CALCULATION OF C-BRACKETS

A. Exact Formulas

1. General, no repeated C_i:

$$\left[C_1, C_2, \ldots, C_n\right] = \sum_{i=1}^{n} \frac{e^{-C_i}}{\prod\limits_{\substack{j=1 \\ j\neq i}}^{n} (C_j - C_i)}$$

2. General, r-fold C_m:

$$\left[C_1, C_2, \ldots, \overbrace{C_m, C_m, C_m}^{r}, \ldots, C_n\right] = \sum_{\substack{i=1 \\ i\neq m}}^{n} \frac{e^{-C_i}}{\prod\limits_{\substack{j=1 \\ j\neq i}}^{n} (C_j - C_i)} + e^{-C_m} \sum_{k=1}^{r} \frac{\phi^{(r-k)}(-C_m)}{(r-k)!(k-1)!}$$

where $\phi^{(r-k)}(-C_m)$ is the (r-k)th derivative with respect to α of the function

$$\phi(\alpha) = \frac{1}{\prod\limits_{\substack{i=1 \\ i\neq m}}^{n} (\alpha + C_i)}$$

evaluated at $\alpha = -C_m$. For example, take the case of 2-fold C_m in a 3-C'd bracket $\left[C_1, C_1, C_3\right]$. The first term becomes

$$\frac{e^{-C_3}}{(C_1 - C_3)(C_1 - C_3)} .$$

The second term is

$$\phi(\alpha) = \frac{1}{\alpha + C_3} \text{ and } \phi(-C_m) = \phi(-C_1) = \frac{1}{C_3 - C_1}$$

$$\phi'(\alpha) = -\frac{1}{(\alpha + C_3)^2} \text{ and } \phi'(-C_1) = -\frac{1}{(C_3 - C_1)^2}$$

so the entire second term becomes

$$e^{-C_1}\left[\frac{1}{(C_3-C_1)} - \frac{1}{(C_3-C_1)^2}\right]$$

$$\text{thence } \left[C_1,C_1,C_3\right] = \frac{e^{-C_3} + e^{-C_1}\left[C_3-C_1-1\right]}{(C_3-C_1)^2} .$$

3. r-fold C_m, no other factors:

$$\Big[\overbrace{C_m,C_m,C_m, \ldots ,C_m}^{r}\Big] = \frac{e^{-C_m}}{(r-1)!}$$

$$\text{and the special case } \Big[\overbrace{0,0, \ldots ,0}^{r}\Big] = \frac{1}{(r-1)!}$$

4. $\left[C_1,C_2, \ldots ,C_n\right] = e^{-B}\left[(C_1-B),(C_2-B), \ldots ,(C_n-B)\right]$

B. Approximation Formulas

When some or all of the C_i's are very nearly the same (as in a very short bombardment), the numerical calculation by equation (A-1), in the preceding section, becomes liable to error, there being a very small difference of large terms.* A few infinite series expansions found useful by us in such cases are set down below. Their derivations are to be found in Appendix II.

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*It is usually necessary in a numerical calculation of the C-bracket by equation (A-1) to carry out the operations to several decimal places. Even though the individual C's themselves may be very approximate, in order to obtain a significant answer it is often necessary to use five-place (or more) exponentials from tables and to carry out multiplication and division with a calculating machine, retaining many decimal places.

1. $$\left[C_1, C_2, \ldots, C_n\right] = \left[\frac{1}{(n-1)!} - \frac{\sum_{i=1}^{n} C_i}{n!} + \ldots\right]$$

or more rapidly converging by application of (A-4) with $B = C_{average} = C_i/n$.

2. $$\left[C_1, C_2, \ldots, C_n\right] = e^{-C_{avg}} \left[\frac{1}{(n-1)!} + \left(\frac{\Sigma_2}{(n+1)!} + \frac{\Sigma_3}{(n+2)!} + \ldots \frac{\Sigma_n}{(2n-1)!}\right) + \left(\frac{\Sigma_2^2}{(n+3)!} + 2\frac{\Sigma_2 \Sigma_3}{(n+4)!} + \frac{\Sigma_3^2}{(n+5)!}\right) - \left(\frac{\Sigma_2^3}{(n+5)!} + \ldots\right) + \ldots\ldots\right]$$

where $\Sigma_2 = \sum_{\substack{ij \\ i \neq j}} F_i F_j$, the sum of the products of all combinations of F's, taken two at a time;

$\Sigma_3 = \sum_{\substack{ijk \\ i \neq j \neq k \neq i}} F_i F_j F_k$, the sum of products of all combinations of F's, three at a time, etc.,

and where we define $F_i = C_i - C_{average}$. (See Appendix II for derivation and general terms.)

3. When some of the C_i's are very large, it is often convenient to simplify the calculation of the brackets by the following series:

$$\left[C_1,C_2,C_3,\ldots,C_n\right] = \frac{1}{C_n}\left[C_1,\ldots,C_{n-1}\right] - \frac{1}{C_n^{\,2}}\left[C_1,\ldots,C_{n-1}\right]' + \frac{1}{C_n^{\,3}}\left[C_1,\ldots,C_{n-1}\right]''$$

$$- \cdots + \frac{(-1)^k}{C_n^{\,k+1}}\left[C_1, \ldots ,C_{n-1}\right]^{(k)} + \cdots^{*}$$

where we define the mth derivative of a C-bracket $\left[C_1,C_2, \ldots ,C_n\right]$ by the inverse transform:

$$\left[C_1,C_2,C_3, \ldots ,C_n\right]^{(m)} = \frac{1}{t^{n-m-1}} \mathcal{L}^{-1} \frac{s^m}{\prod\limits_{1\leq i\leq n}\left(s + \frac{C_i}{t}\right)} ;$$

in the case of non-repeated C_i's, this is easily found as:

$$\left[C_1,C_2,C_3,\ldots,C_n\right]^{(m)} = \left[\frac{d^m}{d\beta^m}\sum_{i=1}^{n}\frac{e^{-C_i\beta}}{\prod\limits_{\substack{j=1\\ j\neq i}}^{n}(C_j-C_i)}\right]_{\beta=1} = (-1)^m \sum_{i=1}^{n}\frac{C_i^{\,m}\, e^{-C_i}}{\prod\limits_{\substack{j=1\\ j\neq i}}^{n}(C_j-C_i)} .$$

If $C_n \gg 1$, we often, as a first approximation, neglect all but the first term in the series.

---

*See Appendix III. An interesting corollary to the differentiation process for C-brackets is the following:

$$\left[C_1,C_2, \ldots ,C_n,0\right]' = \left[C_1,C_2, \ldots ,C_n\right],$$

or any number of zeros are removable, one for each differentiation. This relation follows from the second of the series formulas developed in Appendix III.

EXAMPLE: CALCULATION OF YIELDS IN CHAINS OF SUCCESSIVE NUCLEAR TRANSFORMATIONS

As an example, consider the bombardment of a sample of Mo^{95} in a pile and evaluate yields of Mo^{96}, Mo^{97}, and Mo^{98} in terms of ratio of each to the amount of parent Mo^{95} initially present for the total neutron fluxes chosen; make a log-log plot of yield ratio of each vs. total flux. That is, letting $Mo^{95}_{(o)}$ represent the number of atoms of parent Mo^{95} initially present, calculate

$$Mo^{95}_{(t)}/Mo^{95}_{(o)},\ Mo^{96}_{(t)}/Mo^{95}_{(o)},\ Mo^{97}_{(t)}/Mo^{95}_{(o)},\ Mo^{98}_{(t)}/Mo^{95}_{(o)}$$

where $Mo^{N}_{(t)}$ represents the number of atoms of Mo of atomic weight N at a specified time t, or, in this case, at the total neutron fluxes specified.

Calculations will be made at total fluxes of 4×10^{21}, 4×10^{22}, and 4×10^{23} neutrons/cm^2.

As described early in the text, the quantities or constants with which we are concerned to give the desired yields or successive products were the a_i's and b_i's where

(1) the a_i's control the creation of any isotope A_i from its parent, A_i-1, and, in this case,

$$a_i = I\sigma_i\ ,^{*}$$

where I = flux-rate in neutrons/cm^2 sec

and σ_i = capture cross section in cm^2 for A_i-1,

and (2) the b_i's control the destruction of isotope A_i and, in this case,

$b_i = I\sigma_i\ ,^{*}$ where I is the same as above and

σ_i = total capture cross section in cm^2 for A_i.

---

*In the above example radioactive decay does not enter into the picture; in cases where it is present, see text for including $\lambda_i I$ into the a_i's and b_i's.

Then the a's and b's for the parent and successive isotopes in the problem are as follows:

Isotope	(barns)	a (x 10^{24})	b (x 10^{24})
$A_1 = Mo^{95}$	13.4	---	$b_1 = 13.4$ I
$A_2 = Mo^{96}$	1.0	$a_2 = 13.4$ I	$b_2 = 1.0$ I
$A_3 = Mo^{97}$	2.3	$a_3 = 1.0$ I	$b_3 = 2.3$ I
$A_4 = Mo^{98}$	0.4	$a_4 = 2.3$ I	$b_4 = 0.4$ I

To put these constants into the form to use in the general solution, namely,

$$A_{i(t)}/A_{1(o)} = \prod_{2 \leq i \leq n} q_i \left[C_1, C_2, C_3, \ldots, C_n\right]$$

where $q_i = a_i\, t$ and $C_i = b_i\, t$ and

$$\left[C_1, C_2, \ldots, C_n\right] = \sum_{i=1}^{n} \frac{e^{-C_i}}{\prod_{\substack{j=1 \\ j \neq i}}^{n} (C_j - C_i)}$$

we must make a table of $a_i t$'s and $b_i t$'s. But, as stated above, we are not concerned with t in this particular problem but only with I x t, or total neutron flux (nvt). Therefore, we could have immediately written down the table of q's and C's, which are simply

$$q_i = \sigma_i \times nvt \quad \text{and}$$

$$C_i = \sigma_i \times nvt.$$

Hence, prepare the following table:

Total Flux	4×10^{21} n/cm²		4×10^{22} n/cm²		4×10^{23} n/cm²	
	C	e^{-C}	C	e^{-C}	C	e^{-C}
C_1	$13.4 \times 10^{-24} \times 4 \times 10^{21} = 0.0536$	0.94781	0.536	0.58508	5.36	0.0047
C_2	$1.0 \times 10^{-24} \times 4 \times 10^{21} = 0.0040$	0.99601	0.04	0.96079	0.4	0.67032
C_3	$2.3 \times 10^{-24} \times 4 \times 10^{21} = 0.0092$	0.99084	0.092	0.91211	0.92	0.39852
C_4	$0.4 \times 10^{-24} \times 4 \times 10^{21} = 0.0016$	0.99840	0.016	0.98413	0.16	0.85214
			q		q	
q_2	$13.4 \times 10^{-24} \times 4 \times 10^{21} = 0.0536$		0.536		5.36	
q_3	$1.0 \times 10^{-24} \times 4 \times 10^{21} = 0.004$		0.04		0.4	
q_4	$2.3 \times 10^{-24} \times 4 \times 10^{21} = 0.0092$		0.092		0.92	

We are now ready to calculate the desired yields from the above general solution at the three nvt's chosen; namely, calculate

$$Mo^{95}_{(t)}/Mo^{95}_{(o)} = e^{-C_1};\ Mo^{96}_{(t)}/Mo^{95}_{(o)} = q_2\left[C_1,C_2\right];\ Mo^{97}_{(t)}/Mo^{95}_{(o)} = q_2q_3\left[C_1,C_2,C_3\right]$$

$$\text{and } Mo^{98}_{(t)}/Mo^{95}_{(o)} = q_2q_3q_4\left[C_1,C_2,C_3,C_4\right].$$

The remainder of the example is for the purpose of illustrating the evaluating of the C-brackets, both by the general solution and by alternate equations when indicated. Arithmetic steps have been included for the purpose of illustrating the methods of handling which we have found most practical.

Evaluation of C-brackets at nvt = 4×10^{21} n/cm^2

$\left[C_1, C_2\right]$: The evaluation of a 2-C'd bracket is obviously simply the difference of the exponentials divided by the difference of the C's, so

$$\left[C_1, C_2\right] = 0.0482/0.0496 = 0.972$$

$\left[C_1, C_2, C_3\right]$:

$$\frac{0.94781}{-0.0496 \times -0.0444} + \frac{0.99601}{+0.0496 \times +0.0052} + \frac{0.99084}{+0.0444 \times -0.0052}$$

$$= \frac{+0.94781 \times 0.0052 + 0.99601 \times 0.0444 - 0.99084 \times 0.0496}{0.0496 \times 0.0444 \times 0.0052}$$

$$= \frac{0.0000057}{0.0496 \times 0.0444 \times 0.0052} = 0.495$$

Note that this solution involved differences between very similar numbers, due to C_2 and C_3 being very close together. A check on the C-bracket by Approximation Formula (B-1) might be indicated:

$$\left[C_1, C_2, C_3\right] = \left[1/(3-1)! - (0.0536 + 0.004 + 0.0092)/3!\right]$$

$$= 0.5 - 0.0111 = 0.4889, \text{ showing good agreement.}$$

$\left[C_1, C_2, C_3, C_4\right]$: Inspection shows that C_3 and C_4 are even closer together than the similar C's in the previous bracket, so the rapidly converging form of the infinite series, equation (B-2), will be used.

$$C_1 = 0.0536 \qquad F_1 = +0.0365$$
$$C_2 = 0.0040 \qquad F_2 = -0.0131$$
$$C_3 = 0.0092 \qquad F_3 = -0.0079$$
$$C_4 = 0.0016 \qquad F_4 = -0.0155$$

$$C_{avg} = \frac{0.0684}{4} = 0.0171$$

First term: $e^{-0.0171}/3! = 0.983/6 = 0.164$

Second term: establish $\sum$'s with aid of the table below:

	+0.0365	-0.0131	-0.0079	-0.0155
+0.0365		-0.000479	-0.000288	-0.000566
-0.0131			+0.0001035	+0.000203
-0.0079				+0.0001225
-0.0155				

$$\sum_2 = -0.000904$$

$$\sum_3 = (-0.0131 \times -0.000288) + (-0.0131 \times -0.000566) + (-0.0079 \times -0.000566) + (-0.0155 \times +0.0001035)$$

$$= +0.00000377 + 0.00000742 + 0.00000447 - 0.00000161$$

$$= 1.4 \times 10^{-5}$$

The $\sum$ terms are obviously negligible (and were evaluated only to show the method), thus

$$\left[C_1, C_2, C_3, C_4\right] = 0.164$$

Then, at nvt = 4×10^{21} n/cm^2,

$$Mo^{95}/Mo^{95}_{(o)} = e^{-C_1} = 0.948$$

$$Mo^{96}/Mo^{95}_{(o)} = 0.0536 \times 0.972 = 5.21 \times 10^{-3}$$

$$Mo^{97}/Mo^{95}_{(o)} = 0.0536 \times 0.004 \times 0.495 = 1.06 \times 10^{-4}$$

$$Mo^{98}/Mo^{95}_{(o)} = 0.0536 \times 0.004 \times 0.0092 \times 0.164 = 3.23 \times 10^{-7}$$

Evaluation of C-brackets at remaining nvt's

In the same manner as illustrated above, the C-brackets for nvt of 4×10^{22} and 4×10^{23} n/cm^2 would be calculated. For purposes of illustration, the calculation of the $\left[C_1, C_2, C_3, C_4\right]$ bracket at 4×10^{23} is shown below by two methods, a choice of which is indicated inasmuch as C_4 is becoming large.

(1) $\left[C_1, C_2, C_3, C_4\right]$ at 4×10^{23} n/cm^2 by the general solution A-1 (see table of C and e^{-C} values at beginning of problem):

$$\frac{.0047}{-4.96 \times -4.44 \times -5.2} + \frac{.67032}{+4.96 \times +.52 \times -.24} + \frac{.39852}{+4.44 \times .52 \times -.76} + \frac{.85214}{+5.2 \times .24 \times +.76}$$

$$= \frac{-.0047 \times .24 \times .52 \times .76 - .67032 \times 4.44 \times 5.2 \times .76 + .39852 \times 4.96 \times 5.2 \times .24 + .85214 \times 4.90 \times 4.44 \times .52}{4.96 \times 4.44 \times 5.2 \times .24 \times .52 \times .76}$$

$$= .0427$$

(2) $\left[C_1, C_2, C_3, C_4\right]$ at 4×10^{23} n/cm^2 by use of equation B-3, to be used when some of the C's are large:

$$\left[C_1, C_2, C_3, C_4\right] = \frac{1}{5.36}\left[0.4,\ 0.92,\ 0.16\right] - \frac{1}{5.36^2}\left[0.4,\ 0.92,\ 0.16\right]'$$

First bracket:

$$\frac{0.67032}{+0.52 \times -0.24} + \frac{0.39852}{-0.52 \times -0.76} + \frac{0.85214}{+0.24 \times +0.76} = 0.308$$

Second bracket, illustrating use of the derivative of a bracket, first term only:

$$\left[0.4,\ 0.92,\ 0.16\right]' = -1\left[\frac{0.4e^{-0.4}}{+0.52 \times -0.24} + \frac{0.92e^{-0.92}}{-0.52 \times -0.76} + \frac{0.16e^{-0.16}}{+0.24 \times +0.76}\right]$$

$$= -1\left[\frac{-0.0444}{0.52 \times 0.24 \times 0.76}\right] = +0.474$$

Thus

$$\left[C_1,C_2,C_3,C_4\right] = \frac{0.308}{5.36} - \frac{0.474}{5.36^2} = 0.041$$

which gives good agreement with the first method.

Below is a table of values which were calculated for each isotope at each total flux, following which is a log-log plot of same in Fig. 1.

Table of Ratios of Isotopes to Parent Mo^{95} Initially Present

	Total flux or nvt (n/cm^2)		
Isotope	4×10^{21}	4×10^{22}	4×10^{23}
Mo^{95}	0.948	0.585	4.7×10^{-3}
Mo^{96}	5.21×10^{-2}	0.4	0.72
Mo^{97}	1.06×10^{-4}	8.56×10^{-3}	0.19
Mo^{98}	3.23×10^{-7}	2.78×10^{-4}	8.35×10^{-2}

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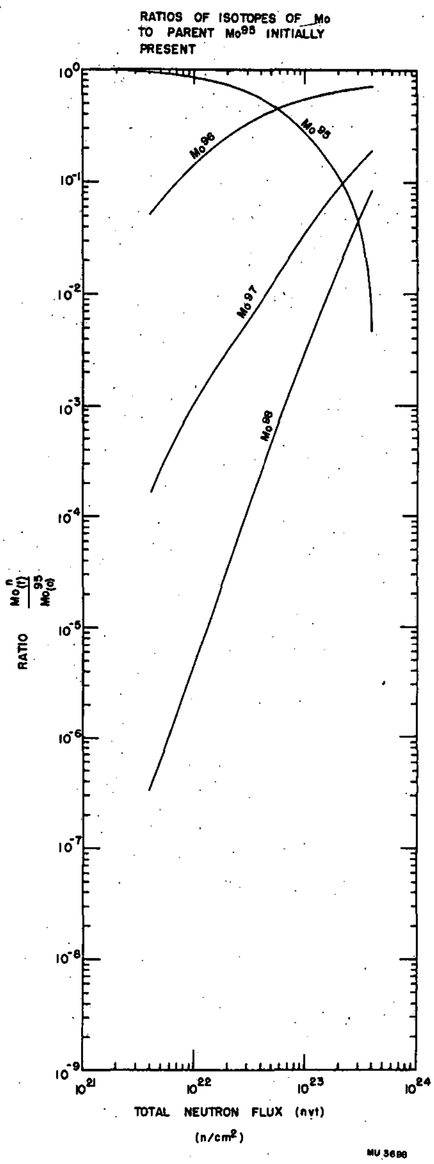

Fig. 1. Yield of molybdenum isotopes in a thermal neutron irradiation of Mo95.

APPENDIX I

GENERAL SOLUTION OF THE EQUATION BY THE LAPLACE TRANSFORM METHOD

The Laplace transform of a function $y(t)$ is defined as

$$Y(s) = \int_0^\infty e^{-st} y(t)\, dt .$$

We multiply both sides of the differential equations of page 5 by e^{-st} and integrate from zero to infinity. This step reduces the set of simultaneous first order linear differential equations into a set of simultaneous first order linear algebraic equations for the transforms. These can be solved by algebraic methods for each transform as a function of the variable s. With the aid of a table of Laplace transforms we can obtain the desired solutions $y_i(t)$.

With the initial conditions at t = 0, as stated above,

$$y_1 = A$$
$$y_i = 0, \quad i \neq 1$$

we apply the Laplace transformation to these equations to reduce them to a series of n linear simultaneous equations in n unknowns.

$$s\,Y_1 - A = -b_1\,Y_1$$
$$s\,Y_2 = a_2 Y_1 - b_2 Y_2$$
$$\cdots\cdots\cdots\cdots$$
$$sY_n = a_n Y_{n-1} - b_n Y_n$$

Rearranging,

$$(s+b_1)Y_1 = A$$
$$(s+b_2)Y_2 = a_2Y_1$$
$$\cdots\cdots\cdots$$
$$(s+b_n)Y_n = a_nY_{n-1}\,.$$

Solving for Y_n, we have

$$(1)\quad Y_n(s) = \frac{A\ a_2a_3\ \ldots\ a_n}{(s+b_1)(s+b_2)\ \ldots\ (s+b_n)} = \frac{A \prod_{2\leq i\leq n} a_i}{\prod_{1\leq i\leq n} (s+b_i)}\,.$$

The solution is obtained by taking the inverse transformation,

$$(2)\quad Y_n(t) = A \prod_{2\leq i\leq n} a_i\, \mathcal{L}^{-1} \frac{1}{\prod_{1\leq i\leq n} (s+b_i)}\,.$$

For convenience in calculation we separate the three factors

$$A;\ t^{n-1} \prod_{2\leq i\leq n} a_i;\ \text{and}\ \frac{1}{t^{n-1}} \mathcal{L}^{-1} \frac{1}{\prod_{1\leq i\leq n} (s+b_i)}$$

multiplying the second and dividing the third factor by t^{n-1} in order to make them dimensionless. For convenience in calculation the substitutions

$$q_i = t\ a_i \ \text{and}\ C_i = t\ b_i$$

are made to introduce the dimensionless parameters q_i and C_i.

The three factors are now

A, the initial amount of isotope A_1 at the head of the chain;

$\prod_{2\leq i\leq n} q_i$, the product of the n-1 production parameters;

and a quantity designated as the "C-bracket," a function of the n C_i's,* or

$$\left[C_1, C_2, \ldots, C_n\right] = \frac{1}{t^{n-1}} \mathcal{L}^{-1} \frac{1}{\prod_{1 \leq i \leq n} (s+b_i)} .$$

The main problem in the calculation lies in the evaluation of the inverse transform or C-bracket factor.

The inverse Laplace transform can always be expressed in an exact manner by the method of splitting up

$$\frac{1}{\prod_{1<i<n} (s+b_i)}$$

into a sum of n-1 partial fractions and taking the inverse transform for each fraction. However, this general solution as a sum of simple exponential functions may not be practical for numerical calculation in some actual cases to be encountered in the experimental work. Good series approximations may often be used to avoid unnecessary complication in numerical calculation of the C-bracket.

The inverse Laplace transform of

$$\frac{1}{\prod_{1 \leq i \leq n} (s+b_i)}$$

is more readily determined by contour integration in the complex plane than by partial fractions.[6] The general contour integral[7] for the inverse transform is as follows (with α a positive real number):

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*Note that the value of the C-bracket is independent of the order of the C_i's. This rearrangement property may be useful in simplifying calculations.

$$\mathcal{L}^{-1} \frac{1}{\prod_{1<i<n} (s+b_i)} = \frac{1}{2\pi i} \int_{\alpha-i\infty}^{\alpha+i\infty} \frac{e^{st}}{\prod_i (s+b_i)} ds$$

The integrand has poles at $s = -b_i$. The integration may be carried out over a closed contour as shown in Fig. 2, if R tends to infinity.

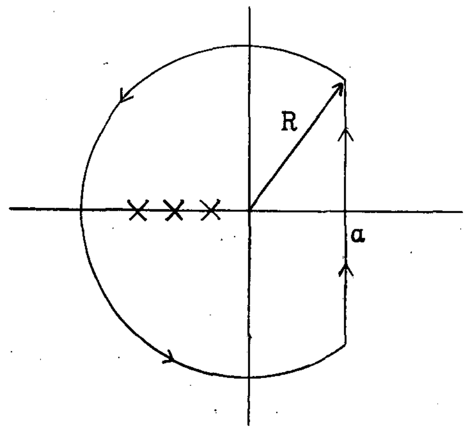

Fig. 2. Contour for integration of the inverse Laplace transform integral.

The contour surrounds all the poles of the integrand, and assuming all poles are simple, the theory of residues gives the value of the integral as

$$\int_{\alpha-i\infty}^{\alpha+i\infty} \frac{e^{st}}{\prod_i (s+b_i)} ds = 2\pi i \sum_k \frac{e^{-b_k t} (s+b_k)}{\prod_i (s+b_i)}$$

and the general solution for the inverse

$$\mathcal{L}^{-1} \frac{1}{\prod_{1\leq i\leq n} (s+b_i)} = \sum_{1\leq k\leq n} \frac{e^{-b_k t} (s+b_k)}{\prod_{1\leq i\leq n} (s+b_i)} .$$

If the poles are not simple, the appropriate formulas for evaluating residues of higher order poles must be applied.

APPENDIX II

DERIVATION OF INFINITE SERIES EXPANSIONS USEFUL FOR SMALL C_i'S

It can be readily seen that an expansion of a Laplace transform as a power series in (1/s) will become on application of the inverse merely a power series in t.

Thus we take the transform of the type encountered here

$$f(s) = \frac{1}{\prod_{i=1}^{n}(s+b_i)} = \frac{1}{s^n \prod_{i=1}^{n}(1+\frac{b_i}{s})}$$

$$= \frac{1}{s^n} \prod_{i=1}^{n}(1+\frac{b_i}{s})^{-1}$$

$$= \frac{1}{s^n}\left[1 + \frac{\sum_{i=1}^{n} b_i}{s} + \frac{\sum_{\substack{ij \\ i\neq j}} b_i b_j}{s^2} + \dots \frac{\prod_{i=1}^{n} b_i}{s^n}\right]^{-1} .$$

But $(1-x)^{-1} = 1 - x + x^2 - x^3 \dots + (-1)^k x^k + \dots$.

$$\text{Let } Q(s) = \left[\frac{\sum b_i}{s} + \frac{\sum b_i b_j}{s^2} + \dots \frac{\prod_{i=1}^{n} b_i}{s^n}\right];$$

$$\text{then } f(s) = \frac{1}{s^n}(1 - Q + Q^2 - Q^3 + \dots).$$

Introduce the notation for the multiple summations taking all combinations of b's:

$$\sum b_i = \sum{}_1; \ \sum b_i b_j = \sum{}_2, \text{ etc.}$$

$\sum_k$ means the sum of all the products of all possible combinations of b_i's taken k at a time. This is a sum of

$$\frac{n!}{k!(n-k)!}$$

products.

Applying the inverse transform to f(s) we have

$$\mathcal{L}^{-1} f(s) = \frac{t^{n-1}}{(n-1)!} - \sum_1 \frac{t^n}{n!} - \sum_2 \frac{t^{n+1}}{(n+1)!} - \dots - \sum_n \frac{t^{2n-1}}{(2n-1)!}$$

$$+ \sum_1^2 \frac{t^{n+1}}{(n+1)!} + \sum_2^2 \frac{t^{n+3}}{(n+3)!} + \sum_3^2 \frac{t^{n+5}}{(n+5)!}$$

$$+ \sum_1 \sum_2 \frac{t^{n+2}}{(n+2)!} \quad \text{etc.}$$

In our definition of the C-bracket the powers of t will all be replaced by unity and the b_i's by dimensionless C_i's as

$$(C_1, \dots ,C_n) = \frac{1}{(n-1)!} - \frac{\sum_1}{n!} - \frac{\sum_2}{(n+1)!} - \dots$$

$$+ \frac{\sum_1^2}{(n+1)!} + \frac{\sum_2^2}{(n+3)!} +$$

$$+ \frac{\sum_1 \sum_2}{(n+2)!} + \dots ,$$

where we understand the $\sum_k$ now as formed from the C_i rather than the b_i.

It is apparent, of course, that the convergence of the series can be hastened by the simple application of transformation (A-4), selecting some sort of average C value for the B.

The general expression of the series can best be given as

$$\frac{1}{t^{n-1}} \mathcal{L}^{-1} f(s) = \frac{1}{(n-1)!} - \frac{1}{t^{n-1}} \mathcal{L}^{-1} \frac{Q}{s^n} + \frac{1}{t^{n-1}} \mathcal{L}^{-1} \frac{Q^2}{s^n} - \dots$$

$$\text{where } Q(s) = \frac{\sum_1}{s} + \frac{\sum_2}{s^2} + \dots + \frac{\sum_n}{s^n} .$$

Note that each term in the general expression will have in its denominator $(n-1+i)!$ where i is the sum of the indices of the $\sum_k$ in the product.

If we introduce the notation,

$$J(i,j,k) = \frac{\sum_i \sum_j \sum_k}{(n-1+i+j+k)!}$$

we can write

$$[C_1, C_2, \dots, C_n] = \frac{1}{(n-1)!} - \sum_{\ell=1} J(\ell) + \sum_{\substack{\ell,m=1 \\ \text{including} \\ \ell=m}} J(\ell,m)$$

$$- \sum_{k} \sum_{\ell=1}^{n} \sum_{m} J(k,\ell,m) + \dots .$$

APPENDIX III

DERIVATION OF INFINITE SERIES EXPANSIONS RAPIDLY CONVERGENT FOR LARGE C_i'S

Taking the transform of the general type for the C-bracket:

$$f(s) = \frac{1}{\prod_{i=1}^{n} (s+b_i)}$$

$$= \frac{1}{(s+b_n) \prod_{i=1}^{n-1} (s+b_i)}$$

$$= \frac{1}{s+b_n} \phi(s) .$$

Now we write:

$$\frac{1}{s+b_n} = \frac{1}{b_n\left(1+\frac{s}{b_n}\right)} = \frac{1}{b_n}\left(1+\frac{s}{b_n}\right)^{-1}$$

$$= \frac{1}{b_n} - \frac{s}{b_n{}^2} + \frac{s^2}{b_n{}^3} - \dots + \frac{\pm(-1)^k s^k}{b_n{}^{k+1}} + \dots .$$

Then

$$f(s) = \frac{1}{b_n} \phi(s) - \frac{1}{b_n{}^2} s\phi(s) + \frac{1}{b_n{}^3} s^2\phi(s) - \dots \frac{(-1)^k s^k \phi(s)}{b_n{}^{k+1}} + \dots .$$

Now the inverse of a transform $\phi(s)$ multiplied by s^k is just the kth derivative of the inverse of $\phi(s)$. $\phi(s)$ is just the transform corresponding to the bracket with C_n not present.

Applying the inverse transform to the above equation and dividing by t^{n-1}, we get the following:

$$\left[C_1, C_2, \ldots, C_n\right] = \frac{1}{C_n}\left[C_1, C_2, \ldots, C_{n-1}\right] - \frac{1}{C_n^{\,2}}\left[C_1, C_2, \ldots, C_{n-1}\right]' + \ldots \frac{+(-1)^k}{C_n^{\,k+1}}\left[C_1, \ldots, C_{n-1}\right]^{(k)} + \ldots ,$$

where the kth derivative is given by

$$\left[C_1, C_2, \ldots, C_{n-1}\right]^{(k)} = \left[\frac{d^k}{d\beta} \sum \frac{e^{-C_1\beta}}{\prod (C_j - C_i)}\right]_{\beta=1} = (-1)^k \sum_{i=1}^{n-1} \frac{C_i^k\, e^{-C_i}}{\prod\limits_{i \neq j} (C_j - C_i)} .$$

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