47. Lundgren, F. A., and Nargolwalla, S. S., *Anal. Chem.*, 40(4), 672 (1968).
48. Burns, F. C., Priest, G. L., and Priest, H. F., *Soc. Appl. Spec.* Chicago, June (1966).
49. Tompson, M. N., and Taylor, J. M., *Nucl. Instr. and Meth.*, 37, 306 (1965).
50. Cochran, R. G., and Henry, K. M., *Rev. Sci. Instr.*, 26, 761 (1955).
51. Downs, W. E., *Tran. Am. Nucl. Soc.*, 10(1), 83 (1967).
52. Evans, R. D., *The Atomic Nucleus*, McGraw-Hill, New York (1967).
53. Zysin, Y. A., Lbov, A. A., and Sel'chenkov, L. I., *Fission Product Yields and their Mass Distribution*, Consultants Bureau, New York (1964).

CHAPTER 5

GROWTH AND DECAY OF RADIOACTIVITY DURING AND AFTER IRRADIATION

I. Laws of Radioactive Decay – Exponential Law

(A) SIMPLE CASE

Consider the case of a radioactive nucleus (1) decaying into a stable nucleus (2):

$$(1) \xrightarrow{\lambda} (2)\ (\text{stab.}) \tag{5.1}$$

This case can be compared to a monomolecular reaction. The reaction rate (number of disintegrations per second, D) is proportional to N the number of atoms of (1) present:

$$\frac{dN}{dt} = -\lambda N = D \tag{5.2}$$

λ is a constant, characteristic of the particular radioactive species, and is called the decay constant, having the dimension of a reciprocal time, t^{-1}. The integral of this simple differential equation is given by

$$N(t) = N^0 \exp(-\lambda t) \tag{5.3}$$

Here, $N(t)$ represents the number of atoms (1) at a time t, N^0 the number at $t = 0$. The radioactive decay is thus governed by an exponential law. Combining (5.2) and 5.3) one can write:

$$\boxed{D(t) = D^0 \exp(-\lambda t)} \tag{5.4}$$

Measuring a fraction z of the real number of disintegrations (i.e. the experimentally observed activity A) one obtains:

$$A = zD = z\lambda N$$

and

$$A(t) = A^0 \exp(-\lambda t) \tag{5.5}$$

The coefficient z is called the detection coefficient and will depend on the nature of the detection instrument, the efficiency for the recording of the particular radiation with that particular instrument, and the geometrical arrangement of sample and detector.

The characteristic "rate of a radioactive decay" may conveniently be given in terms of the half-life $T_{1/2}$, i.e. the time required for an initial (large) number of atoms to be reduced to half that value. Thus, at $t = T_{1/2}$, $N = N^0/2$ and

$$\ln \tfrac{1}{2} = -\lambda T_{1/2} \quad \text{or} \quad T_{1/2} = 0.693/\lambda \tag{5.6}$$

This half-life can conveniently be determined graphically by plotting A *vs.* t on a semilog scale giving a straight line with a slope $-\lambda$ (Figure 5.1).

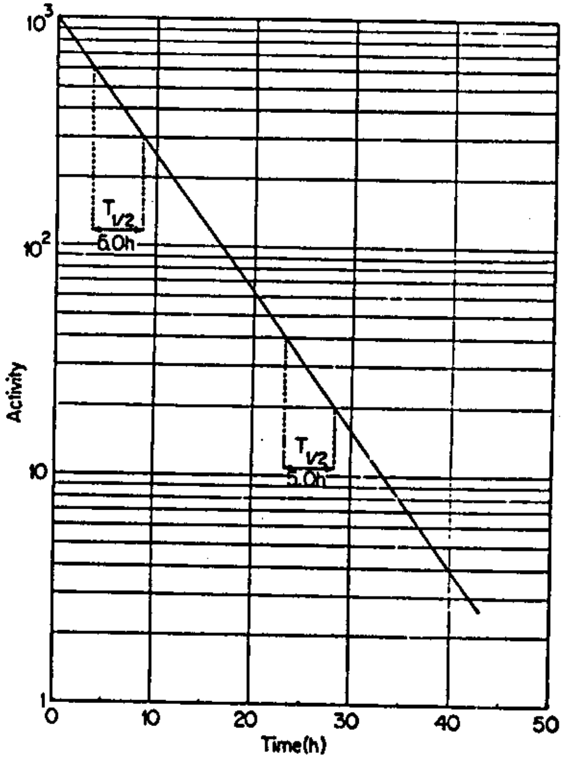


Fig. 5.1. Radioactive decay of a single radionuclide ($T_{1/2}$ = 5.0 h)
$A(t) = A^0 \exp(-\lambda t)$
$\log A(t) = \log A^0 - \lambda t$

(B) Mixtures of Independently Decaying Activities

In the above equations the radioactivity, corresponding to the transformation of a single atomic species, was considered. In the more general case, the observed total activity is obviously the sum of all the independent activities, i.e.

$$A = A_1 + A_2 + \ldots = z_1\lambda_1 N_1 + z_2\lambda_2 N_2 + \ldots \tag{5.7}$$

The detection coefficients $z_1, z_2 \ldots$ can be quite different, depending on the nature and the energy of the radiation of the different species.

For a mixture of several independent activities the result of plotting $\log A$ *vs.* t is always an upward concave curve (Figure 5.2). In the case of two radioactive species one can write:

$$A(t) = A_1(t) + A_2(t) = A_1^0 \exp(-\lambda_1 t) + A_2^0 \exp(-\lambda_2 t) \tag{5.8}$$

Assuming that (1) decays more rapidly than (2), i.e. $\lambda_1 > \lambda_2$, equation (5.8) simplifies to

$$A(t) \approx A_2^0 \exp(-\lambda_2 t) \tag{5.9}$$

after a sufficiently long time. Extrapolating this straight line in the semilog plot to $t \to 0$, one can subtract the value $A_2(t)$ for any time, from the corresponding total activity A; this allows one to find A_1 as a function of t, i.e. $A_1^0 \exp(-\lambda_1 t)$. The complex decay curve is analyzed into its two components, from both of which now, in the usual way (*cf.* Figure 5.2), $T_{1/2}$ may be determined.

Due to experimental uncertainties in the observed data, this procedure is limited to mixtures of only two or three radioisotopes, and even two component curves may not be satisfactorily resolved if the two half-lives differ by less than about a factor of two or if the rates differ by a large factor.

In Figure 5.3 a complex decay curve is analyzed into four components. The accuracy in the determination of the half-lives of the short-lived components is however poor.

During the past years a number of computer methods for analyzing multicomponent radioactive decay curves have been developed. As expected, the earlier codes had rather limited capabilities, while the later modifications became increasingly complex and sophisticated (see Chapter 9, section III, C, 3).

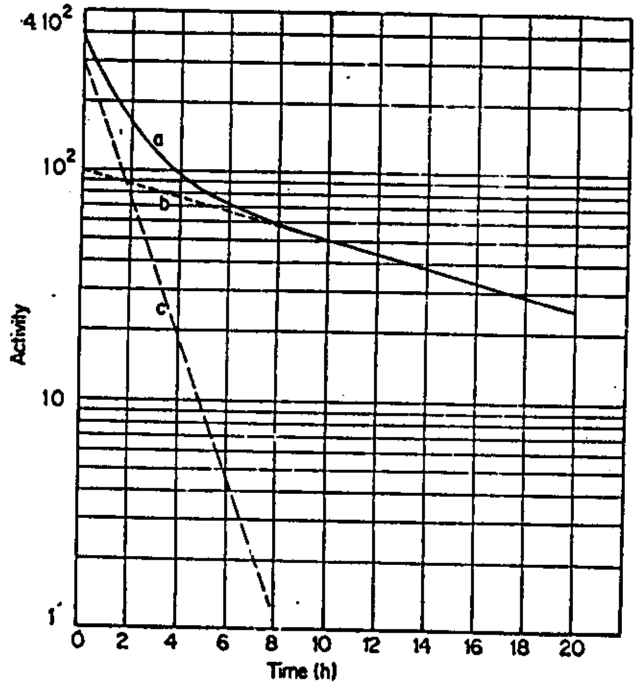


Fig. 5.2. Analysis of composite decay curve (mixture of two independently decaying activities)

a = composite decay curve: $A(t) = A_1^0 \exp(-\lambda_1 t) + A_2^0 \exp(-\lambda_2 t)$
b = longer-lived component: $A_2(t) = A_2^0 \exp(-\lambda_2 t)$
c = shorter-lived component: $A_1(t) = A_1^0 \exp(-\lambda_1 t)$

Normally a least-squares fitting operation is performed (1,2). The data of a radioactive decay curve consist of m measurements of the counting rates A_i of the sample at times t_i. If n independent nuclear species are present, then the set of data satisfies m equations of the form

$$A_i = \sum_{j=1}^{j=n} A_j^0 \exp(-\lambda_j t_i) + Z_i \tag{5.10}$$

where an individual term in the sum, $A_j^0 \exp(-\lambda_j t_i)$ represents the contribution of the jth component in the total activity at time t_i. The residual Z_i at that point is due to statistical fluctuations and experimental errors. Since the n coefficients A_j^0 enter these equations linearly a solution by the least-squares method is possible. The condition for such a solution is that

$$\sum_{i=1}^{i=m} W_i Z_i^2 = \text{minimum} \tag{5.11}$$

where W_i is the weight assigned to the square of each residual ($W_i = 1/\sigma_i^2$). It is convenient to adopt a matrix notation for further calculations. The code is normally intended to be run by the "monitor" system, i.e. all reasonable errors will cause a suitable comment to be printed by the machine before it takes appropriate action.

The data needed to begin the least-squares fitting calculation include: time, counts, length of count, background, and the half-lives for each component. The data obtained from the calculation include: the activity of each component at the time of the first count, the deviation of each point from the calculated curve, weighted by $1/\sigma^2$ for that point and the "error matrix".

On the average the machine resolution of decay curves gives better results than those obtained by hand. Not the least advantage is the

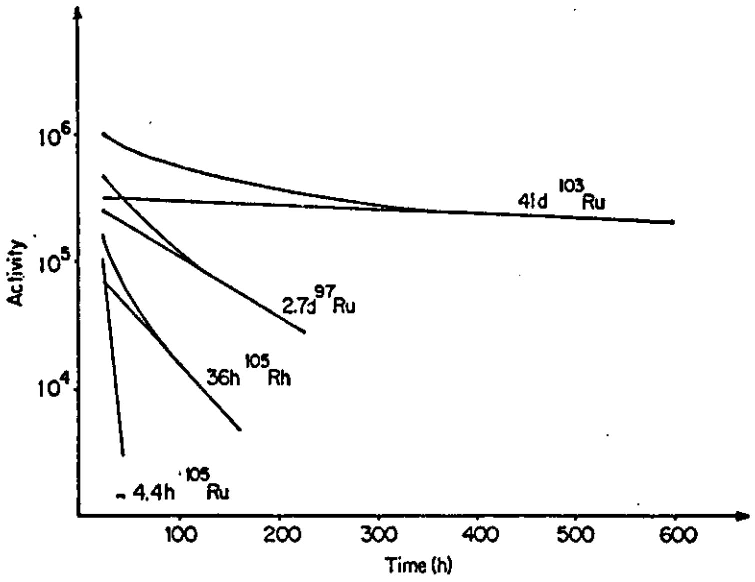


Fig. 5.3. Decay curve of irradiated ruthenium.

speed of the calculation. On an IBM 7090 a typical calculation involving 30 data points and including data read-in and evaluation, resolution of three components, recalculation if necessary, print out of answers and tabulation of collected data will take about ten seconds of machine time.

In none of the above methods does the number of components "fall out" of the analysis and if the number of components is not known, any "accurate" estimate of the parameters begins to lose its meaning. The two essential difficulties inherent in this technique are, that one must deal with data that only approximate $A(t)$ over a finite range in t, and that the exponential series possesses such strongly non orthogonal properties, that the parameters are extremely sensitive to minor fluctuations in the data. The approach of Gardner *et al* (3,4) is based on the fact that the exponential series can be represented by a Laplace integral equation:

$$A(t) = \sum_{i=1}^{n} A_i^0 \exp(-\lambda_i t) = \int_0^\infty g(\lambda) \exp(-\lambda t)\, \mathrm{d}\lambda \qquad (5.12)$$

Here $g(\lambda)$ is a sum of delta functions, but due to the error inherent in the experimental estimate of $A(t)$ and in the numerical computations necessary to obtain $g(\lambda)$, a plot of $g(\lambda)$ *vs.* λ appears in the form of a frequency spectrum. The presence of a true peak in the spectrum indicates a component, the abscissa value at the center of the peak being the decay constant λ_i whereas the height of the peak is proportional to the coefficient A_i^0. The function $g(\lambda)$ is obtained by Fourier transforms, as described in full detail by Gardner *et al* (4).

This method is applicable both to the decay of independent species and also to the case of growth and decay chains. One advantage of the method lies in the fact that it is not necessary to have initial estimates of the parameters (particularly λ_i) before the analysis as in curve fitting procedures, such as the non-linear least-squares method. Furthermore it is an approach where the number of components automatically "falls out" of the analysis. Full use is made of the accuracy inherent in the data since they are treated as a whole, as opposed to some "subtraction type" methods, wherein all but the shortest-lived components are determined using fewer points than are actually available. Finally, the occurrence of very similar λ values does not endanger the entire solution as in other methods.

II. Growth of Radioactive Daughters

(A) Two Successive Decays

Consider the simple case of a radioactive species (1), which decays to produce another radioactive species (2):

$$(1) \xrightarrow{\lambda_1} (2) \xrightarrow{\lambda_2} (3)\ \text{(stable)} \qquad (5.13)$$

Decay of (1) and growth and decay of (2) are described by the following differential equations.

$$\frac{\mathrm{d}N_1}{\mathrm{d}t} = -\lambda_1 N_1 \quad \text{where } N_1 = N_1^0 \exp(-\lambda_1 t) \qquad (5.14)$$

$$\frac{\mathrm{d}N_2}{\mathrm{d}t} = \lambda_1 N_1 - \lambda_2 N_2 \qquad (5.15)$$

as the second species is formed at the rate at which the first decays $(\lambda_1 N_1)$ and decays at a rate $-\lambda_2 N_2$.

From (5.14) or (5.3) and (5.15) it follows that

$$\frac{\mathrm{d}N_2}{\mathrm{d}t} + \lambda_2 N_2 = \lambda_1 N_1^0 \exp(-\lambda_1 t) \qquad (5.16)$$

This is a linear differential equation of the first order. The general form of this so-called Leibnitz equation is given by

$$\frac{\mathrm{d}y}{\mathrm{d}x} + Py = Q \quad \text{(Leibnitz equation)} \qquad (5.17)$$

where P and Q can be functions of x and explicitly independent of y, or constant. As this equation is of great importance, it will be treated in detail. The general solution is given by:

$$y = \exp\left(-\int P\,\mathrm{d}x\right)\int Q \exp\left(\int P\,\mathrm{d}x\right)\mathrm{d}x + \mathrm{C}.\exp\left(-\int P\,\mathrm{d}x\right) \qquad (5.18)$$

In the case of (5.16): $y = N_2$, $x = t$, $P = \lambda_2$, $Q = \lambda_1 N_1^0 \exp(-\lambda_1 t)$. Thus

$$N_2(t) = \exp(-\lambda_2 t) \int_0^t \lambda_1 N_1^0 \exp(-\lambda_1 t) \exp(\lambda_2 t)\,\mathrm{d}t + \mathrm{C} \exp(-\lambda_2 t)$$

$$N_2(t) = \exp(-\lambda_2 t)\, \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} [\exp(\lambda_2 - \lambda_1)t - 1] + \mathrm{C} \exp(-\lambda_2 t)$$

$$N_2(t) = \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1} [\exp(-\lambda_1 t) - \exp(-\lambda_2 t)] + \mathrm{C} \exp(-\lambda_2 t)$$

Finally, if $t = 0$, $N_2^0 = 0 + C$, so $C = N_2^0$.

The final solution is thus given by

$$\boxed{N_2(t) = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 [\exp(-\lambda_1 t) - \exp(-\lambda_2 t)] + N_2^0 \exp(-\lambda_2 t)} \tag{5.19}$$

Equation (5.19) is the general expression for the decay of a radionuclide, formed from another radionuclide.

The last term gives the contribution at any time from the daughter atoms present initially. Assuming that, for $t = 0$, $N_2^0 = 0$ (the case of a daughter activity, growing in the freshly purified parent fraction), equation (5.19) is simpified to

$$N_2(t) = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 [\exp(-\lambda_1 t) - \exp(-\lambda_2 t)] \tag{5.20}$$

In equation (5.19) and (5.20) two general cases can be distinguished, depending on which of the two substances (parent or daughter) has the longer half-life. This discussion will be limited to equation (5.20), i.e. assuming that $N_2^0 = 0$.

1. *Transient Equilibrium*

$\lambda_1 < \lambda_2$, e.g. $(T_{1/2})_1 = 5.0$ h and $(T_{1/2})_2 = 0.5$ h, i.e. $\lambda_2 = 10\lambda_1$.

Consider first the number of parent atoms as a function of time. At $t = 0$, $N_1 = N_1^0$ and the corresponding activity $A_1^0 = \lambda_1 N_1^0$ (e.g. $A_1^0 = 90$ cpm).

Hence, as a function of time, one can write $N_1 = N_1^0 \exp(-\lambda_1 t)$ or $A_1 = A_1^0 \exp(-\lambda_1 t)$ (Figure 5.4 curve aa), assuming $z_1 = 1$.

To calculate the daughter activity in the parent-plus-daughter fraction, equation (5.20) must be applied. After t becomes sufficiently large, $\exp(-\lambda_2 t)$ is negligible compared to $\exp(-\lambda_1 t)$, hence:

$$N_2 \approx \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 \exp(-\lambda_1 t) \quad \text{and} \quad A_2 \approx \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} N_1^0 \exp(-\lambda_1 t)$$

assuming $z_2 = 1$. Substituting $\lambda_2 = 10\lambda_1$, this becomes:

$$A_2 \approx \frac{10}{9} \lambda_1 N_1^0 \exp(-\lambda_1 t) \quad \text{(Figure 5.4 curve bb)}$$

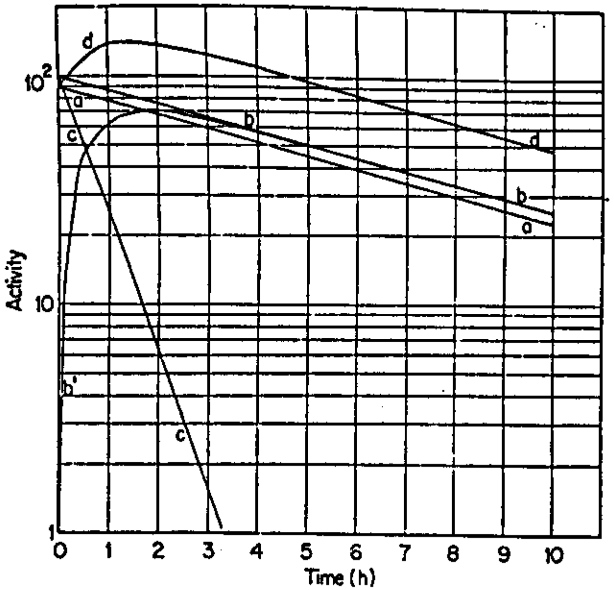


Fig. 5.4. Transient equilibrium; $(T_{1/2})_1 = 5.0$ h and $(T_{1/2})_2 = 0.5$ h; $(\lambda_2 = 10\lambda_1)$

curve aa: activity due to parent
curve bb: daughter activity in the parent-plus-daughter fraction
curve b'bb: daughter activity growing in freshly purified parent fraction
curve cc: decay of freshly isolated daughter fraction
curve dd: total activity of an initially pure parent fraction.

i.e. the daughter activity in the parent-plus-daughter fraction decays with the half-life of the parent. Extrapolating this straight line in the semilog plot to $t \to 0$, one finds

$$\frac{10}{9} \lambda_1 N_1^0 = 100 \text{ cpm},$$

since $A_1^0 = 90$ cpm.

For small values of t, the general equation (5.20) has to be applied to calculate N_2 or A_2; actually, (5.20) can be written as the algebraic sum of two exponential terms:

$$A_2 = \frac{10}{9} \lambda_1 N_1^0 \exp(-\lambda_1 t) - \frac{10}{9} \lambda_1 N_1^0 \exp(-\lambda_2 t).$$

The first term is represented by curve bb (Figure 5.4). The second term which has the same extrapolated value as the first one

$$\left(\frac{10}{9}\lambda_1 N_1^0 \text{ at } t = 0\right),$$

describes the decay of a freshly isolated daughter fraction (separated from the parent), and is represented by curve cc.

Thus, curve bb minus curve cc (= curve bb′) represents the daughter activity in the parent-plus-daughter fraction as a function of time. At $t = 0$, $A_2 = 0$. After t becomes sufficiently large, A_2 = curve bb; between these extreme values, there is a continuous transition.

The total activity of an initially pure parent fraction is given by

$$A = A_1 + A_2 = \lambda_1 N_1^0 \exp(-\lambda_1 t) + \frac{10}{9}\lambda_1 N_1^0 \exp(-\lambda_1 t) - \frac{10}{9}\lambda_1 N_1^0 \exp(-\lambda_2 t)$$

or, graphically: curve aa plus curve bb′ = curve dd (Figure 5.4) e.g. at $t = 0$, $A^0 = A_1^0 + A_2^0 = A_1^0 = 90$ cpm; at $t = 5.0$ h, $A = 45 + 50 = 95$ cpm.

Remark: The figure assumes $z_1 = z_2 = 1$ and $N_2^0 = 0$ (initially pure parent fraction).

Conclusions for transient equilibrium. After t becomes sufficiently large, the daughter activity and the total activity decay with the half-life of the parent. The daughter can, of course, only decay after being formed; so, its decay rate is determined by its formation; the latter is equal to the decay rate of the parent.

In these conditions, the ratio "number of atoms of parent to number of atoms of daughter" is constant.

$$\frac{N_2}{N_1} = \frac{\lambda_1}{\lambda_2 - \lambda_1} \qquad (5.21)$$

Multiplying both numbers of this equation with $z_2\lambda_2/z_1\lambda_1$, one finds an analogous equation:

$$\frac{A_2}{A_1} = \frac{z_2\lambda_2 N_2}{z_1\lambda_1 N_1} = \frac{z_2\lambda_2}{z_1(\lambda_2 - \lambda_1)}$$

or

$$\frac{z_1 A_2}{z_2 A_1} = \frac{\lambda_2}{\lambda_2 - \lambda_1} \qquad (5.22)$$

Notice, however, that the right-hand sides of equations (5.21) and (5.22) are not the same.

In the case of the example ($z_1 = z_2$, $\lambda_2 = 10\lambda_1$), this becomes

$$\frac{A_2}{A_1} = \frac{10\lambda_1}{10\lambda_1 - \lambda_1} = \frac{10}{9}$$

and

$$\frac{A_2 + A_1}{A_1} = \frac{A}{A_1} = \frac{19}{9} \quad \text{or} \quad \frac{A_2 + A_1}{A_2} = \frac{A}{A_2} = \frac{19}{10}$$

which can be verified on Figure 5.4.

2. *Secular Equilibrium*

If $\lambda_1 \ll \lambda_2$, one has a limiting case of radioactive equilibrium and the same considerations can be made as in II, A, 1. It is called secular equilibrium. Setting $(T_{1/2})_1 = \infty$ and $(T_{1/2})_2 = 1.0$ h, the number of parent atoms is given by

$$N_1 = N_1^0 \exp(-\lambda_1 t) \approx N_1^0 \; (\approx \text{constant})$$

as λ_1 is very small. The parent activity is given by

$$A_1 \approx A_1^0 = \lambda_1 N_1^0 = \text{constant} = R \text{ (Figure 5.5 curve aa)}$$

i.e. the parent activity does not decrease measurably during many daughter half-lives.

After t becomes large compared to $(T_{1/2})_2$, the daughter activity in the parent-plus-daughter fraction can be calculated from

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 \exp(-\lambda_1 t) \approx \frac{R}{\lambda_2} \text{ and } A_2 \approx R$$

(in Figure 5.5, same curve aa as for parent activity). Extrapolating this constant to $t \to 0$, one finds of course, R.

For small values of t, the general equation (5.17) must be applied to calculate N_2 or A_2:

$$N_2 = \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1}[\exp(-\lambda_1 t) - \exp(-\lambda_2 t)] \approx \frac{R}{\lambda_2}[1 - \exp(-\lambda_2 t)] \qquad (5.23)$$

and

$$A_2 = R[1 - \exp(-\lambda_2 t)] \text{ or } A_2 = R - R\exp(-\lambda_2 t)$$

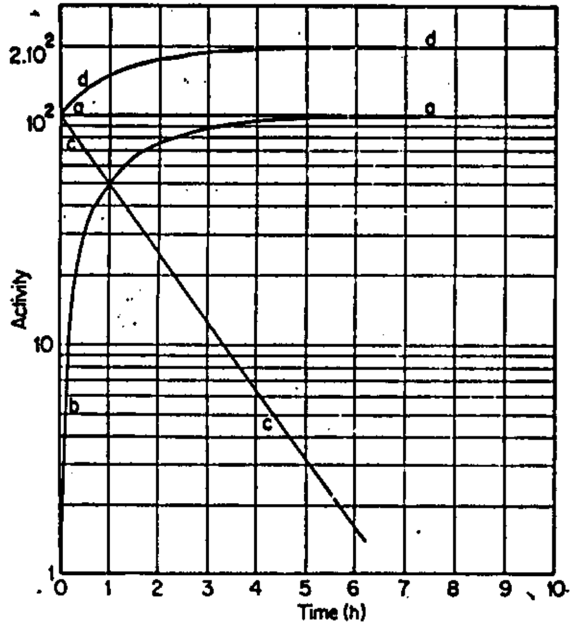


Fig. 5.5. Secular equilibrium; $(T_{1/2})_1 = \infty$ and $(T_{1/2})_2 = 1.0$ h
curve aa: activity due to parent
curve cc: decay of freshly isolated daughter fraction
curve ba: daughter activity growing in freshly purified parent fraction
curve dd: total activity in an initially pure parent fraction.

Thus, one can conclude that A_2 is given by the difference of a constant term R (Figure 5.5 curve aa) and an exponential term $R \exp(-\lambda_2 t)$. The latter term, which has the same extrapolated value as the first one (R at $t = 0$), describes the decay of a freshly isolated daughter fraction and is represented by curve cc.

$$\text{Curve aa minus curve cc} = \text{curve ba} = A_2.$$

At $t = 0$, $A_2 = 0$; for t sufficiently large, A_2 = curve aa $= R$.

The total activity is calculated as follows:

$$A = A_1 + A_2 = R + R - R \exp(-\lambda_2 t)$$

i.e. curve aa plus curve ba gives curve dd.

At $t = 0$: $A = R + R - R = R$ $(= A_1)$; for t sufficiently large: $A = R + R - 0 = 2R$.

Between these extreme values, there is a continuous transition. Figure 5.5 assumes $z_1 = z_2 = 1$ and $N_2^0 = 0$.

3. *Case of no equilibrium*

$\lambda_1 > \lambda_2$, e.g. $(T_{1/2})_1 = 0.5$ h and $(T_{1/2})_2 = 5.0$ h, i.e. $\lambda_1 = 10\lambda_2$.

Further assume: $z_1 = z_2 = 1$ and $N_2^0 = 0$.

The number of parent atoms as a function of time is given by

$$N_1 = N_1^0 \exp(-\lambda_1 t)$$

and the corresponding activity by

$$A_1 = \lambda_1 N_1^0 \exp(-\lambda_1 t) \text{ (Figure 5.6 curve aa; } A_1^0 = 900 \text{ cpm)}$$

The daughter activity in the parent-plus-daughter fraction is again calculated from equation (5.20). After t becomes sufficiently large, this equation is simplified to:

$$N_2 = \frac{\lambda_1}{\lambda_1 - \lambda_2} N_1^0 \exp(-\lambda_2 t) \quad \text{and} \quad A_2 = \frac{\lambda_1 \lambda_2}{\lambda_1 - \lambda_2} N_1^0 \exp(-\lambda_2 t)$$

In the case of the numerical example, this becomes:

$$A_2 = \frac{10}{9} \lambda_2 N_1^0 \exp(-\lambda_2 t) \quad \text{(Figure 5.6 curve bb).}$$

This means: the daughter decays with its own half-life. Extrapolating this straight line in the semilog plot to $t \to 0$, one finds

$$\frac{10}{9} \lambda_2 N_1^0 = \frac{\lambda_1 N_1^0}{9} = \frac{A_1^0}{9} = 100 \text{ cpm.}$$

For smaller values of t, the general equation (5.20) must be used; A_2 is given by the difference of two exponential terms:

$$A_2 = \frac{\lambda_1 \lambda_2}{\lambda_1 - \lambda_2} N_1^0 [\exp(-\lambda_2 t) - \exp(-\lambda_1 t)]$$

$$A_2 = \frac{\lambda_1 N_1^0}{9} \exp(-\lambda_2 t) - \frac{\lambda_1 N_1^0}{9} \exp(-\lambda_1 t)$$

The first term is represented by curve bb (Figure 5.6). The second term, which has the same extrapolated value as the first one (100 cpm

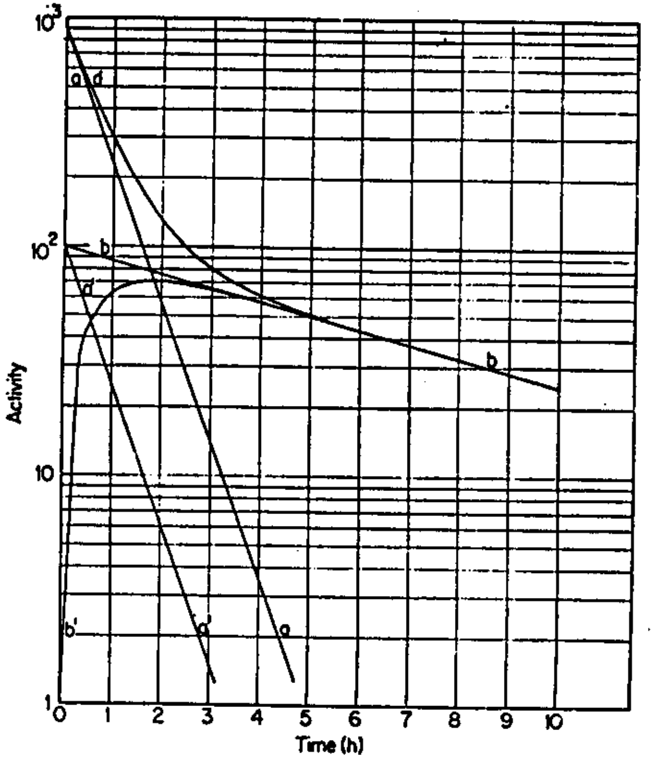


Fig. 5.6. Case of no equilibrium $(T_{1/2})_1 = 0.5$ h and $(T_{1/2})_2 = 5.0$ h; $(\lambda_1 = 10\lambda_2)$.
curve aa: activity due to parent
curve bb: daughter activity in the parent-plus-daughter fraction
curve b′b: daughter activity growing in freshly purified parent fraction
curve db: total activity in an initially pure parent fraction.

at $t = 0$) is 1/9 $(=\lambda_2/(\lambda_1 - \lambda_2))$ of curve aa, and is represented by curve a′a′. Curve bb minus curve a′a′ = curve b′b = A_2.

At $t = 0$, $A_2 = 0$; after t becomes sufficiently large, A_2 = curve bb. Between these extreme values, there is a continuous upward concave curve.

The total activity is given by:

$$A = A_1 + A_2 = \text{aa} + \text{b'b} = \text{db}$$

At $t = 0$, $A^0 = A_1^0 = 900$ cpm; after t becomes sufficiently large, $A = A_2$ (curve bb).

Remark: Observing in practice the total activity of an initially pure parent fraction, one obtains in a semilog plot a curve as db. After a sufficient time, the longer-lived daughter activity entirely dominates and its half life, $(T_{1/2})_2 = 5.0$ h, may be read from this portion of the decay curve.

Extrapolating this straight portion to $t \to 0$, one finds

$$„A_2^{0}" = \frac{\lambda_1\lambda_2}{\lambda_1 - \lambda_2} N_1^0 \quad \text{or better} \quad \frac{z_2\lambda_1\lambda_2}{\lambda_1 - \lambda_2} N_1^0.$$

Extrapolating the first part of the total decay curve to $t \to 0$, one gets

$$A_1^0 = \lambda_1 N_1^0 \quad \text{or better} \quad z_1\lambda_1 N_1^0.$$

From these two equations follows:

$$\frac{A_1^0}{„A_2^{0}"} = \frac{z_1\lambda_1 N_1^0(\lambda_1 - \lambda_2)}{z_2\lambda_1\lambda_2 N_1^0} = \frac{z_1}{z_2}\cdot\frac{\lambda_1 - \lambda_2}{\lambda_2} \tag{5.24}$$

If $\lambda_1 \gg \lambda_2$ this reduces to

$$\frac{A_1^0}{„A_2^{0}"} = \frac{z_1}{z_2}\cdot\frac{\lambda_1}{\lambda_2} = \frac{z_1}{z_2}\cdot\frac{(T_{1/2})_2}{(T_{1/2})_1} \tag{5.25}$$

i.e. graphical interpretation of the total decay curve enables one not only to find $(T_{1/2})_2$, but also $(T_{1/2})_1$, provided z_1 and z_2 are known.

In the numerical example one finds $(T_{1/2})_2 = 5.0$ h (see semilog plot, Figure 5.6). $A_1^0/„A_2^{0}" = 900/100 = 9$, thus $(\lambda_1 - \lambda_2)/\lambda_2 = 9$; $\lambda_1 - \lambda_2 = 9\lambda_2$; $\lambda_1 = 10\lambda_2$ (if $z_1 = z_2$) or $(T_{1/2})_1 = 0.5$ h.

(B) MANY SUCCESSIVE DECAYS

Consider a series, where transformation from one member to another occurs by radioactive decay,

$$(1) \xrightarrow{\lambda_1} (2) \xrightarrow{\lambda_2} (3) \xrightarrow{\lambda_3} \ldots (n) \xrightarrow{\lambda_n} \tag{5.26}$$

The rates of nuclide transformation are described by the following set of differential equations:

$$\frac{dN_1}{dt} = -\lambda_1 N_1$$

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \qquad (5.27)$$

$$\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3 \text{ etc.}$$

where $N_1(t)$ and $N_2(t)$ are given by equations (5.3) and (5.19) respectively. Substituting (5.19) in the third differential equation, $N_3(t)$ can be calculated. This substitution method of the i^{th} differential equation into the $(i + 1)^{\text{th}}$ is, however, rather cumbersome. H. Bateman (5) has given the solution for a chain of n members, assuming that at $t = 0$ the parent fraction alone is present, i.e. $N_2^0 = N_3^0 = \ldots N_n^0 = 0$. This solution can be formulated as follows:

$$\boxed{N_n(t) = \lambda_1\lambda_2\lambda_3 \ldots \lambda_{n-1} N_1^0 \sum_{i=1}^{i=n} C_i \exp(-\lambda_i t)} \qquad (5.28)$$

where:

$$\boxed{C_i = \prod_{j=1}^{j=n} \frac{1}{\lambda_j - \lambda_i} \, (i \neq j)} \qquad (5.29)$$

Example 1.

Case number (5.13), i.e. a chain of only two members, can of course be treated in the same way, with the special assumption that $N_2^0 = 0$.

$$C_1 = \frac{1}{\lambda_2 - \lambda_1}; \quad C_2 = \frac{1}{\lambda_1 - \lambda_2}$$

$$N_2(t) = \lambda_1 N_1^0 \left[\frac{\exp(-\lambda_1 t)}{\lambda_2 - \lambda_1} + \frac{\exp(-\lambda_2 t)}{\lambda_1 - \lambda_2}\right]$$

$$= \frac{\lambda_1}{\lambda_2 - \lambda_1}[N_1^0 \exp(-\lambda_1 t) - \exp(-\lambda_2 t)]$$

This is identical to equation (5.20).

Example 2.

Calculate N_3 as a function of time for a chain of three members, assuming that $N_2^0 = N_3^0 = 0$ at $t = 0$.

Applying equations (5.28) and (5.29), one finds:

$$C_1 = \frac{1}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}; \; C_2 = \frac{1}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)};$$

$$C_3 = \frac{1}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)}$$

$$N_3(t) = \lambda_1\lambda_2 N_1^0 \left[\frac{\exp(-\lambda_1 t)}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} + \frac{\exp(-\lambda_2 t)}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} + \frac{\exp(-\lambda_3 t)}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)}\right]$$

Remark: If a solution is required for the more general case with N_2^0, $N_3^0 \ldots N_n^0 \neq 0$, one can construct it by adding to the Bateman solution for N_n in an n-membered chain, a Bateman solution for N_n in an $(n - 1)$-membered chain with species 2 as the parent (thus $N_2 = N_2^0$ at $t = 0$ replaces N_1^0 in equation (5.28)), and an analogous Bateman solution for N_n in an $(n - 2)$-membered chain with species 3 as the parent ($N_3 = N_3^0$ at $t = 0$ replaces N_1^0 in equation (5.28)), etc.

(C) BRANCHING DECAY

Consider a radionuclide (1), decaying in two different ways forming radionuclides (2) and (2′) respectively.

$$\begin{tikzcd}
 & (2') \arrow[r] & (3') \\
(1) \arrow[ur, "\lambda_1'"] \arrow[dr, "\lambda_1"'] & & \\
 & (2) \arrow[r] & (3)
\end{tikzcd} \qquad (5.30)$$

λ_1 and λ_1' are partial decay constants, related to the formation of (2) and (2′) respectively; (2) is formed at a rate $\lambda_1 N_1$; (2′) at a rate $\lambda_1' N_1$; (1) decays at a rate $(\lambda_1 + \lambda_1')N_1$.

Applying the Bateman equation to such a chain, one must replace equation (5.28) by

$$N_n(t) = \lambda_1^*\lambda_2^* \ldots \lambda_{n-1}^* N_1^0 \sum_{i=1}^{i=n} C_i \exp(-\lambda_i t) \qquad (5.31)$$

i.e., the λ's before $\sum$ must be replaced by partial decay constants (λ^*): λ^* is the decay constant for the transformation of the i^{th} chain member to the $(i + 1)^{\text{th}}$ one. In the chain (1)–(2′)–(3′) (5.30), λ_1^* is equal to λ_1', but λ_1 (5.31) = $(\lambda_1 + \lambda_1')$ (5.30).

If a decay chain branches and subsequently the two branches are *joined*, the two branches are treated by this method as separate chains; the production of a common member beyond the branch point is the sum of the number of atoms from the two paths. (See for instance III, C, 2 and III, D, 2).

III. Transformation in a Neutron Flux

These transformation equations will be stated for the case of a neutron flux. They are, of course, equally applicable for any other activating particle.

(A) SIMPLE CASE

Consider first the simple case of a transformation in a neutron flux, represented by the scheme

$$(1) \xrightarrow[(n,\gamma)]{\sigma_1} (2) \xrightarrow[\beta^-]{\lambda_2} (3)\ \text{(stable)} \tag{5.32}$$

Example: ${}^{23}\text{Na} \xrightarrow[(n,\gamma)]{0.54\ \text{b}} {}^{24}\text{Na} \xrightarrow[\beta^-]{15.0\ \text{h}} {}^{24}\text{Mg}$ (stable).

Thus, stable nuclei (1) are activated by neutron capture (cross section σ_1) and transformed into radioactive nuclei (2), decaying to the stable nuclei (3) (decay constant λ_2).

1. *Solution of the Leibnitz Equation*

During irradiation, the growth of nuclei (2) is proportional to the activation cross section σ_1, to the neutron flux φ and to the number of atoms N_1 (thus $\varphi\sigma_1 N_1$); one can assume that in most cases N_1 remains constant during the irradiation, i.e. that the burn-up is negligible ($N_1 \approx N_1^0$). On the other hand, the radioactive nuclei (2) decay during the irradiation process at a rate $\lambda_2 N_2$. Consequently the following differential equation can be formulated:

$$\frac{\mathrm{d}N_2}{\mathrm{d}t_b} = \varphi\sigma_1 N_1^0 - \lambda_2 N_1 \tag{5.33}$$

This equation is of the general form (5.17), where $x = t_b$ (irradiation time), $P(x) = \lambda_2$ = constant, $Q(x) = \varphi\sigma_1 N_1^0$ = constant, and $y = N_2$.

The solution is given by:

$$N_2(t_b) = \exp(-\lambda_2 t_b) \int_0^{t_b} \varphi\sigma_1 N_1^0 \exp(\lambda_2 t_b)\, \mathrm{d}t_b + C \exp(-\lambda_2 t_b)$$

$$N_2(t_b) = \frac{\varphi\sigma_1 N_1^0}{\lambda_2} \exp(-\lambda_2 t_b)\,[\exp(\lambda_2 t_b) - 1] + C \exp(-\lambda_2 t_b)$$

$$N_2(t_b) = \frac{\varphi\sigma_1 N_1^0}{\lambda_2} [1 - \exp(-\lambda_2 t_b)] + C \exp(-\lambda_2 t_b)$$

Finally, if $t_b = 0$, $N_2^0 = 0 + C$, hence $C = N_2^0$.

So, the final solution is given by

$$N_2(t_b) = \frac{\varphi\sigma_1 N_1^0}{\lambda_2} [1 - \exp(-\lambda_2 t_b)] + N_2^0 \exp(-\lambda_2 t_b) \tag{5.34}$$

If at $t_b = 0$, $N_2^0 = 0$, equation (5.34) is simplified to

$$N_2(t_b) = \frac{\varphi\sigma_1 N_1^0}{\lambda_2} [1 - \exp(-\lambda_2 t_b)] \tag{5.35}$$

The disintegration rate of the nuclei (2) as a function of the irradiation time is given by:

$$\boxed{D_2(t_b) = \lambda_2 N_2(t_b) = \varphi\sigma_1 N_1^0 [1 - \exp(-\lambda_2 t_b)]} \tag{5.36}$$

The factor $1 - \exp(-\lambda_2 t_b)$ is called saturation factor S. For $t_b \gg (T_{1/2})_2$, $\exp(-\lambda_2 t_b) \to 0$ and $S \to 1$; hence D_2 reaches a maximum. For $t_b \ll (T_{1/2})_2$, $\exp(-\lambda_2 t_b) \approx 1 - \lambda_2 t_b$ and $S \approx \lambda_2 t_b$, i.e. the activity increases proportionally to t_b. The disintegration rate after an irradiation time t_b and a waiting time t can be calculated, by substituting (5.33) in (5.4), setting $t_0 = t_b$:

$$\boxed{D_2(t_b, t) = D_2(t_b) \cdot \exp(-\lambda_2 t) = \varphi\sigma_1 N_1^0 [1 - \exp(-\lambda_2 t_b)] \cdot \exp(-\lambda_2 t)} \tag{5.37}$$

(see Figure 5.7).

2. *Solution by the Generalized Bateman Equation*

The problem can also be solved by means of a generalized Bateman equation (5.28), as proposed by Rubinson (6). For that purpose, a new

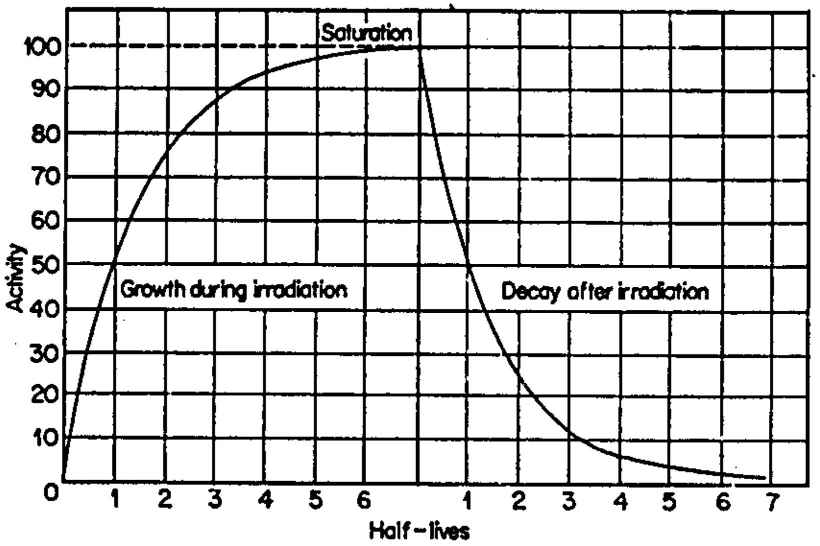


Fig. 5.7. Growth and decay of radioactivity
$A_2(t_b) = \varphi\sigma_1 N_1^0[1 - \exp(-\lambda_2 t_b)]$
$A_2(t_b, t) = \varphi\sigma_1 N_1^0 [1 - \exp(-\lambda_2 t_b)] \exp(-\lambda_2 t)$

modified "disappearance constant" is introduced: $\Lambda = \lambda + \varphi\sigma$. If one considers N atoms of a radioactive species, with a decay constant λ (s^{-1}) and a total (effective) reaction cross section σ (cm^2) in a constant neutron flux φ ($cm^2\ s^{-1}$), the disappearance of this species is no longer governed by radioactive decay alone ($\lambda N\ s^{-1}$); the disappearance by the transmutation reaction must also be taken into account ($\varphi\sigma N\ s^{-1}$). So, the total rate of disappearance is given by $(\lambda + \varphi\sigma)N\ s^{-1} = \Lambda N\ s^{-1}$ (setting $\lambda + \varphi\sigma = \Lambda$). Λ can be considered as a modified "disappearance constant". If a given nuclide disappears by a nuclear reaction only, $\Lambda = \varphi\sigma$; if a given radionuclide disappears by radioactive decay only, $\Lambda = \lambda$.

If one considers a chain in which the transformation from one number to the next occurs by a nuclear reaction and by radioactive decay, equations (5.28) or (5.31) can be applied, provided that λ_i or λ_i^* is replaced by $\Lambda_i = \lambda_i + \varphi\sigma_i$ or $\Lambda_i^* = \lambda_i^* + \varphi\sigma_i^*$, where the asterisks serve as a reminder that in either branching decay or branching activation λ_i^* is the partial decay constant and σ_i^* is the partial reaction cross section.

Assuming $N_2^0 = N_3^0 = \dots N_n^0 = 0$, the solution is given by:

$$\boxed{N_n(t_b) = \Lambda_1^*\Lambda_2^* \dots \Lambda_{n-1}^* N_1^0 \sum_{i=1}^{i=n} C_i \exp(-\Lambda_i t_b)} \tag{5.38}$$

where

$$\boxed{C_i = \prod_{j=1}^{j=i} \frac{1}{\Lambda_j - \Lambda_i} \quad (j \neq i)} \tag{5.39}$$

Applying this procedure for the simple case (5.32) one finds:

$$\Lambda_1 = \Lambda_1^* = \varphi\sigma_1$$
$$\Lambda_2 = \lambda_2$$

Thus

$$C_1 = \frac{1}{\Lambda_2 - \Lambda_1} = \frac{1}{\lambda_2 - \varphi\sigma_1}$$

$$C_2 = \frac{1}{\Lambda_1 - \Lambda_2} = -\frac{1}{\lambda_2 - \varphi\sigma_1}$$

Hence:

$$N_2(t_b) = \Lambda_1^* N_1^0 [C_1 \exp(-\Lambda_1 t_b) + C_2 \exp(-\Lambda_2 t_b)]$$

$$N_2(t_b) = \frac{\varphi\sigma_1 N_1^0}{\lambda_2 - \varphi\sigma_1} [\exp(-\varphi\sigma_1 t_b) - \exp(-\lambda_2 t_b)]$$

In practically all cases (flux and cross section not extremely high, irradiation time not extremely long) one has $\lambda_2 \gg \varphi\, \sigma_1$. Consequently the last equation can be simplified to:

$$N_2(t_b) = \frac{\varphi\sigma_1 N_1^0}{\lambda_2} [1 - \exp(-\lambda_2 t_b)]$$

corresponding to equation (5.35). The disintegration rate after a waiting time t is already given in equation (5.37).

(B) Growth of a Radioactive Daughter in a Neutron Flux

Another case of transformation in a neutron flux, which is also of practical interest, is represented by the scheme:

$$(1) \xrightarrow[(n,\gamma)]{\sigma_1} (2) \xrightarrow[\beta^-]{F_2\lambda_2} (3) \xrightarrow[\beta^-]{\lambda_3} (4)\ \text{(stable)} \tag{5.40}$$

where F_2 is the fraction of disintegrations of nuclides (2) which produces nuclides (3).

Example:

$$^{198}\mathrm{Pt} \xrightarrow[(n,\gamma)]{3.9\,b} {}^{199}\mathrm{Pt} \xrightarrow[\beta^-]{\overset{F_2=1}{31\,m}} {}^{199}\mathrm{Au} \xrightarrow[\beta^-]{3.15\,d} {}^{199}\mathrm{Hg}\ (\text{stable})$$

Indeed, half-lives, nature of the radiation, chemical properties, etc. make it sometimes preferable to measure the daughter (3) activity for the determination of element (1), instead of the activity of nuclide (2). The determination of platinum from ^{199}Au is a typical example.

Calculations of this case will be given in two ways: 1. by describing the rates of nuclide transformation by differential equations of the general form (5.17), and substituting the solution of the first differential equation into the second, etc.; 2. by applying directly the Bateman-Rubinson equations.

1. *Solution of a Set of Leibnitz Equations*

The growth of the number of nuclides (3) as a function of irradiation time t_b is formulated by the following differential equation:

$$\frac{\mathrm{d}N_3}{\mathrm{d}t_b} = F_2\lambda_2N_2 - \lambda_3N_3 \tag{5.41}$$

where $N_2(t_b)$ is given by equation (5.35). Substituting (5.35) into (5.41) gives:

$$\frac{\mathrm{d}N_3}{\mathrm{d}t_b} = F_2\varphi\sigma_1N_1^0[1 - \exp(-\lambda_2t_b)] - \lambda_3N_3$$

This is of the general form (5.17), with $x = t_b$, $y = N_3$, $P(x) = \lambda_3 =$ constant, $Q(x) = F_2\varphi\sigma_1N_1^0[1 - \exp(-\lambda_2t_b)]$.

Integration gives

$$N_3(t_b) = \exp(-\lambda_3t_b)\left[F_2\varphi\sigma_1N_1^0\int_0^{t_b}\exp(\lambda_3t_b)\,\mathrm{d}t_b - F_2\varphi\sigma_1N_1^0\int_0^{t_b}\exp(-\lambda_2t_b)\exp(\lambda_3t_b)\,\mathrm{d}t_b\right] + N_3^0\exp(-\lambda_3t_b)$$

$$N_3(t_b) = F_2\varphi\sigma_1N_1^0\exp(-\lambda_3t_b)\left[\frac{\exp(\lambda_3t_b)-1}{\lambda_3} - \frac{\exp(\lambda_3-\lambda_2)t_b - 1}{\lambda_3-\lambda_2}\right] + N_3^0\exp(-\lambda_3t_b)$$

$$N_3(t_b) = \frac{F_2\varphi\sigma_1N_1^0}{\lambda_3}\left\{[1 - \exp(-\lambda_3t_b)] - \frac{\lambda_3}{\lambda_3-\lambda_2}[\exp(-\lambda_2t_b) - \exp(-\lambda_3t_b)]\right\} + N_3^0\exp(-\lambda_3t_b) \tag{5.42}$$

$$N_3(t_b) = \frac{F_2\varphi\sigma_1N_1^0}{\lambda_3(\lambda_3-\lambda_2)}\{\lambda_3[1 - \exp(-\lambda_2t_b)] - \lambda_2[1 - \exp(-\lambda_3t_b)]\} \tag{5.43}$$

if $N_3^0 = 0$.

For the disintegration rate of radionuclides (3) as a function of irradiation time, the following equation holds:

$$\boxed{D_3(t_b) = \frac{F_2\varphi\sigma_1N_1^0}{\lambda_3-\lambda_2}\{\lambda_3[1 - \exp(-\lambda_2t_b)] - \lambda_2[1 - \exp(-\lambda_3t_b)]\}} \tag{5.44}$$

2. *Solution by the Bateman–Rubinson Equation*

The same result can be obtained by application of equations (5.38) and (5.39).

$\Lambda_1 = \Lambda_1^* = \varphi\sigma_1$
$\Lambda_2 = \lambda_2$ (presuming that nuclides (2) are not activated)
$\Lambda_2^* = F_2\lambda_2$
$\Lambda_3 = \lambda_3$ (presuming that nuclides (3) are not activated).

Hence:

$$C_1 = \frac{1}{(\lambda_2-\varphi\sigma_1)(\lambda_3-\varphi\sigma_1)} \approx \frac{1}{\lambda_2\lambda_3}$$

$$C_2 = \frac{1}{(\varphi\sigma_1-\lambda_2)(\lambda_3-\lambda_2)} \approx \frac{1}{\lambda_2(\lambda_2-\lambda_3)}$$

$$C_3 = \frac{1}{(\varphi\sigma_1-\lambda_3)(\lambda_2-\lambda_3)} \approx \frac{1}{\lambda_3(\lambda_3-\lambda_2)}$$

Simplification was applied, assuming that burn-up is negligible ($\lambda \gg \varphi\sigma$).

Thus, if $N_2^0 = N_3^0 = 0$ one can substitute these values into equation (5.38).

$$N_3(t_b) = F_2\varphi\sigma_1 N_1^0 \lambda_2 \left[\frac{\exp(-\varphi\sigma_1 t_b)}{\lambda_2\lambda_3} + \frac{\exp(-\lambda_2 t_b)}{\lambda_2(\lambda_2-\lambda_3)} - \frac{\exp(-\lambda_3 t_b)}{\lambda_3(\lambda_2-\lambda_3)}\right]$$

$$N_3(t_b) = \frac{F_2\varphi\sigma_1 N_1^0}{\lambda_3}\left\{\exp(-\varphi\sigma_1 t_b) + \frac{1}{\lambda_2-\lambda_3}[\lambda_3 \exp(-\lambda_2 t_b) - \lambda_2 \exp(-\lambda_3 t_b)]\right\}$$

$$N_3(t_b) = \frac{F_2\varphi\sigma_1 N_1^0}{\lambda_3(\lambda_2-\lambda_3)}\{\lambda_2[\exp(-\varphi\sigma_1 t_b) - \exp(-\lambda_3 t_b)] + \lambda_3[\exp(-\lambda_2 t_b) - \exp(-\varphi\sigma_1 t_b)]\}$$

As normally $\exp(-\varphi\sigma_1 t_b) \approx 1$ (burn-up negligible), this result is identical to equation (5.43).

For the calculation of the number of atoms N_3 at any given time t after the end of the irradiation, one must use the general equation for the decay of a radionuclide which produces a radioactive daughter, i.e. equation (5.19); for this particular application $t_0 = t_b$, thus $N_3^0 = N_3(t_b)$, $N_2^0 = N_2(t_b)$.

Hence, for the scheme (5.40), equation (5.19) must be written in the following way:

$$N_3(t) = \frac{\lambda_2}{\lambda_3-\lambda_2} N_2(t_b)[\exp(-\lambda_2 t) - \exp(-\lambda_3 t)] + N_3(t_b)\exp(-\lambda_3 t)$$

where $N_2(t_b)$ and $N_3(t_b)$ are given by equations (5.35) and (5.43). Consequently, $N_3(t_b, t)$ can be calculated by substitution of (5.35) and (5.43) in the above equation. The assumption $F_2 = 1$ will be made here.

$$N_3(t_b, t) = \frac{\varphi\sigma_1 N_1^0}{\lambda_3-\lambda_2}[1-\exp(-\lambda_2 t_b)][\exp(-\lambda_2 t) - \exp(-\lambda_3 t)] + \frac{\varphi\sigma_1 N_1^0}{\lambda_3(\lambda_3-\lambda_2)}\{\lambda_3[1-\exp(-\lambda_2 t_b)] - \lambda_2[1-\exp(-\lambda_3 t_b)]\}\exp(-\lambda_3 t)$$

$$N_3(t_b, t) = \frac{\varphi\sigma_1 N_1^0}{\lambda_3(\lambda_3-\lambda_2)}\{\lambda_3[1-\exp(-\lambda_2 t_b)][\exp(-\lambda_2 t) - \exp(-\lambda_3 t)] + \lambda_3[1-\exp(-\lambda_2 t_b)]\exp(-\lambda_3 t) - \lambda_2[1-\exp(-\lambda_3 t_b)]\exp(-\lambda_3 t)\}$$

$$N_3(t_b, t) = \frac{\varphi\sigma_1 N_1^0}{\lambda_3(\lambda_3-\lambda_2)}\{\lambda_3[1-\exp(-\lambda_2 t_b)]\exp(-\lambda_2 t) - \lambda_2[1-\exp(-\lambda_3 t_b)]\exp(-\lambda_3 t)\} \quad (5.45)$$

The determination of an element by measuring the activity of the daughter (3) is especially useful, if (2) is shorter-lived than (3), i.e. $\lambda_2 \gg \lambda_3$. Besides, (2) is generally irradiated to saturation in that case, i.e. $\exp(-\lambda_2 t_b) \to 0$ and $[1 - \exp(-\lambda_2 t_b)] \to 1$. If the waiting time is long, so as to allow the nuclei (2) to decay completely into nuclei (3), one can simplify the above equation:

$$N_3(t_b, t) \approx \frac{\varphi\sigma_1 N_1^0}{\lambda_3(-\lambda_2)}\{\lambda_3 \exp(-\lambda_2 t) - \lambda_2[1-\exp(-\lambda_3 t_b)]\exp(-\lambda_3 t)\}$$

$$N_3(t_b, t) \approx \frac{\varphi\sigma_1 N_1^0}{\lambda_3}[1-\exp(-\lambda_3 t_b)]\exp(-\lambda_3 t) \quad (5.46)$$

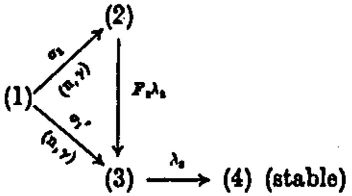
Or

$$\boxed{D_3(t_b, t) \approx \varphi\sigma_1 N_1^0 [1-\exp(-\lambda_3 t_b)]\exp(-\lambda_3 t)} \quad (5.47)$$

This corresponds to equation (5.37).

(C) BRANCHING ACTIVATION

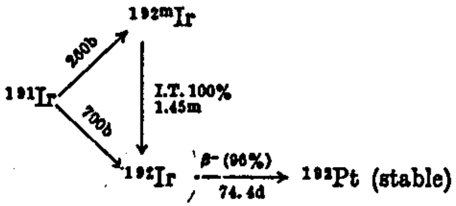
Another frequently occurring case is represented by the following activation scheme:



(5.48)

i.e. the radionuclide (3) is either formed directly or from an isomer (2).

Example:



Again, calculations will be given in two different ways.

1. *Solution of a Set of Leibnitz Equations*

The growth of the number N_3 of nuclides (3) as a function of irradiation time is described by the following differential equation:

$$\frac{\mathrm{d}N_3}{\mathrm{d}t_b} = F_2\lambda_2 N_2 + \varphi\sigma_1' N_1^0 - \lambda_3 N_3 \tag{5.49}$$

where N_2 is given by equation (5.35). Substitution of (5.35) in (5.49) again yields a differential equation of the general type (5.17):

$$\frac{\mathrm{d}N_3}{\mathrm{d}t_b} + \lambda_3 N_3 = F_2\varphi\sigma_1 N_1^0 [1 - \exp(-\lambda_2 t_b)] + \varphi\sigma_1' N_1^0$$

with $x = t_b$, $y = N_3$, $P(x) = \lambda_3 = \text{const.}$, and $Q(x) =$ second term of the above equation.

Assuming $N_2^0 = N_3^0 = 0$, one gets after integration between 0 and t_b:

$$N_3(t_b) = \exp(-\lambda_3 t_b)\varphi N_1^0 \left[\sigma_1 F_2 \int_0^{t_b} \exp(\lambda_3 t_b)\,\mathrm{d}t_b - \sigma_1 F_2 \int_0^{t_b} \exp(\lambda_3 - \lambda_2)t_b\,\mathrm{d}t_b + \sigma_1' \int_0^{t_b} \exp(\lambda_3 t_b)\,\mathrm{d}t_b\right]$$

$$N_3(t_b) = \varphi N_1^0 \exp(-\lambda_3 t_b)\left\{\frac{F_2\sigma_1[\exp(\lambda_3 t_b) - 1]}{\lambda_3} - \frac{F_2\sigma_1[\exp(\lambda_3 - \lambda_2)t_b - 1]}{\lambda_3 - \lambda_2} + \frac{\sigma_1'[\exp(\lambda_3 t_b) - 1]}{\lambda_3}\right\}$$

Thus:

$$D_3(t_b) = \varphi N_1^0 \left\{(F_2\sigma_1 + \sigma_1')[1 - \exp(-\lambda_3 t_b)] + \frac{F_2\sigma_1\lambda_3}{\lambda_2 - \lambda_3}[\exp(-\lambda_2 t_b) - \exp(-\lambda_3 t_b)]\right\} \tag{5.50}$$

2. *Solution by the Bateman–Rubinson Equation*

The method of Rubinson can be applied as well. In this case, the same remark holds as the one given in section II, C, for the Bateman equation (branching decay): if a decay chain (transformation chain) branches and subsequently the two branches join they are treated as separate chains. The production of a common member beyond the branching point is the sum of nuclides from the two paths.

These two chains are respectively:

$$(1) \xrightarrow{\sigma_1} (2) \xrightarrow{F_2\lambda_2} (3) \xrightarrow{\lambda_3} \quad \text{and} \quad (1) \xrightarrow{\sigma_1'} (3) \xrightarrow{\lambda_3}$$

So, $N_3(t_b)$ must be calculated for each of the two chains, and after that, the results are summed. The solution for the first chain is given by equation (5.43) or (5.42), for the second chain by equation (5.35) where λ_2 has to be replaced by λ_3, and σ_1 by σ_1'.

$$N_3(t_b) = \frac{F_2\varphi\sigma_1 N_1^0}{\lambda_3}\left\{[1 - \exp(-\lambda_3 t_b)] - \frac{\lambda_3}{\lambda_3 - \lambda_2}[\exp(-\lambda_2 t_b) - \exp(-\lambda_3 t_b)]\right\} + \frac{\varphi\sigma_1' N_1^0}{\lambda_3}[1 - \exp(-\lambda_3 t_b)]$$

This is, of course, equivalent to equation (5.50).

In practice the isomeric form (2) is generally shorter-lived than (3), i.e. $\lambda_2 \gg \lambda_3$ (e.g. ^{192m}Ir, ^{192}Ir). Moreover, (2) is usually irradiated to saturation in that case ($t_b \gg (T_{1/2})_2$) or $\exp(-\lambda_2 t_b) \to 0$. Assuming that (2) has completely decayed into (3) after a sufficient waiting time, one can simplify equation (5.50):

$$N_3(t_b) \approx \frac{\varphi N_1^0}{\lambda_3}\left\{(F_2\sigma_1 + \sigma_1')[1 - \exp(-\lambda_3 t_b)] - \frac{F_2\sigma_1\lambda_3}{\lambda_2}\exp(-\lambda_3 t_b)\right\}$$

$$N_3(t_b) \approx \frac{\varphi N_1^0}{\lambda_3}\left[F_2\sigma_1 + \sigma_1' - F_2\sigma_1 \exp(-\lambda_3 t_b) - \sigma_1' \exp(-\lambda_3 t_b) - F_2\sigma_1 \frac{\lambda_3}{\lambda_2}\exp(-\lambda_3 t_b)\right]$$

$$N_3(t_b) \approx \frac{\varphi N_1^0}{\lambda_3}\left[F_2\sigma_1 + \sigma_1' - \left(F_2\sigma_1 + F_2\sigma_1\frac{\lambda_3}{\lambda_2} + \sigma_1'\right)\exp(-\lambda_3 t_b)\right]$$

$$N_3(t_b) \approx \frac{\varphi N_1^0}{\lambda_3}(F_2\sigma_1 + \sigma_1')[1 - \exp(-\lambda_3 t_b)] \tag{5.51}$$

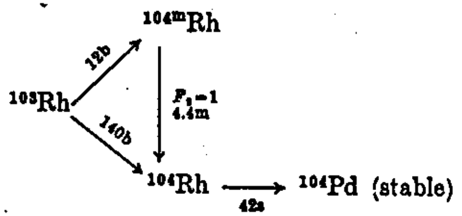
because $\sigma_1 \dfrac{\lambda_3}{\lambda_2} \ll \sigma_1$.

The disintegration rate of nuclide (3) as a function of irradiation time is then given by

$$\boxed{D_3(t_b) \approx \varphi N_1^0 (F_2\sigma_1 + \sigma_1')[1 - \exp(-\lambda_3 t_b)]} \qquad (5.52)$$

which has the same form as (5.36).

In other cases is $\lambda_2 \ll \lambda_3$ and usually $t_b \gg (T_{1/2})_2 \gg (T_{1/2})_3$, e.g.



So, other simplifications in equation (5.50) are possible: $\exp(-\lambda_2 t_b)$ and $\exp(-\lambda_3 t_b) \to 0$:

$$\boxed{D_3(t_b) \approx (F_2\sigma_1 + c_1')\varphi N_1^0} \qquad (5.53)$$

Radioactive decay gives rise to a parent–daughter relation, so that $D_3(t_b, t)$ can be calculated by substitution of (5.50) and (5.36) in (5.19). The latter equation must, in this case, be written in the following way:

$$N_3(t_b, t) = \frac{\lambda_2}{\lambda_3 - \lambda_2} N_2(t_b)[\exp(-\lambda_2 t) - \exp(-\lambda_3 t)]$$

$$+ N_3(t_b)\exp(-\lambda_3 t)$$

$$N_3(t_b, t) = \frac{\varphi N_1^0 \sigma_1}{\lambda_3 - \lambda_2}[1 - \exp(-\lambda_2 t_b)][\exp(-\lambda_2 t) - \exp(-\lambda_3 t)]$$

$$+ \varphi N_1^0 \left\{ \frac{(\sigma_1 + \sigma_1')[1 - \exp(-\lambda_3 t_b)]}{\lambda_3} \right.$$

$$\left. - \frac{\sigma_1[\exp(-\lambda_2 t_b) - \exp(-\lambda_3 t_b)]}{\lambda_3 - \lambda_2} \right\} \exp(-\lambda_3 t)$$

assuming that $F_2 = 1$. Or

$$D_3(t_b, t) = \varphi N_1^0 \left\{ \frac{\sigma_1 \lambda_3}{\lambda_3 - \lambda_2}[1 - \exp(-\lambda_2 t_b)][\exp(-\lambda_2 t) - \exp(-\lambda_3 t)] \right.$$

$$+ (\sigma_1 + \sigma_1')[1 - \exp(-\lambda_3 t_b)]\exp(-\lambda_3 t)$$

$$\left. - \frac{\sigma_1 \lambda_3}{\lambda_3 - \lambda_2}[\exp(-\lambda_2 t_b) - \exp(-\lambda_3 t_b)]\exp(-\lambda_3 t_b) \right\} \qquad (5.54)$$

Again, two important cases will be considered: first $\lambda_2 \gg \lambda_3$, and $\exp(-\lambda_2 t_b) \to 0$ (isomeric form short-lived and irradiated to saturation). This leads to:

$$D_3(t_b, t) \approx \varphi N_1^0 \left\{ \frac{\sigma_1 \lambda_3}{\lambda_2}\exp(-\lambda_3 t) \right.$$

$$+ (\sigma_1 + \sigma_1')[1 - \exp(-\lambda_3 t_b)]\exp(-\lambda_3 t)$$

$$\left. - \frac{\sigma_1 \lambda_3}{\lambda_2}\exp(-\lambda_3 t_b)\exp(-\lambda_3 t) \right\}$$

$$D_3(t_b, t) \approx \varphi N_1^0 . \exp(-\lambda_3 t) \left\{ \sigma_1 \frac{\lambda_3}{\lambda_2} - \sigma_1 \frac{\lambda_3}{\lambda_2}\exp(-\lambda_3 t_b) \right.$$

$$\left. + (\sigma_1 + \sigma_1')[1 - \exp(-\lambda_3 t_b)] \right\}$$

$$D_3(t_b, t) \approx \varphi N_1^0 . \exp(-\lambda_3 t) \left\{ \sigma_1 \frac{\lambda_3}{\lambda_2}[1 - \exp(-\lambda_3 t_b)] \right.$$

$$\left. + (\sigma_1 + \sigma_1')[1 - \exp(-\lambda_3 t_b)] \right\}$$

$$D_3(t_b, t) \approx \varphi N_1^0 . \exp(-\lambda_3 t)[1 - \exp(-\lambda_3 t_b)]\left(\sigma_1 + \sigma_1 \frac{\lambda_3}{\lambda_2} + \sigma_1'\right)$$

If $\lambda_2 \gg \lambda_3$, then $\sigma_1 + \sigma_1 \frac{\lambda_3}{\lambda_2} \approx \sigma_1$ and:

$$\boxed{D_3(t_b, t) \approx (\sigma_1 + \sigma_1')\varphi N_1^0 [1 - \exp(-\lambda_3 t_b)]\exp(-\lambda_3 t)} \qquad (5.55)$$

which has the same form as equation (5.37).

If, however, $\lambda_3 \ll \lambda_2$ and $\exp(-\lambda_2 t_b) \to 0$ and $\exp(-\lambda_3 t_b) \to 0$, as is the case for ^{103}Rh(n, γ), the following simplification can be made:

$$\boxed{D_3(t_b, t) \approx \varphi N_1^0 [\sigma_1 \exp(-\lambda_2 t) + (\sigma_1 + \sigma_1') \exp(-\lambda_3 t)]} \tag{5.56}$$

From the foregoing it is possible to conclude that, in the case of "isomeric activation" ($\lambda_2 \gg \lambda_3$ and $t_b \gg 0.693/\lambda_2$), the activation cross sections $F_2\sigma_1 + \sigma_1'$ must be summed when calculating the activity of radionuclides (3) as a function of irradiation and waiting time. (See equations (5.52) and (5.53)).

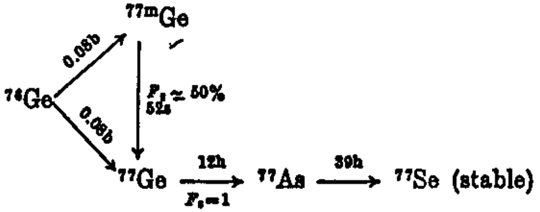
Example: The total activity of ^{192}Ir must be calculated with $\sigma_1' + F_2\sigma_1 = 700b + 260b = 960b$ as cross section, and $0.693/74.4d$ in the saturation factor (equations (5.52)).

(D) GROWTH OF A RADIOACTIVE DAUGHTER AFTER BRANCHING ACTIVATION

Consider the case represented by the following activation scheme:

(5.57)

Example:



Again, calculation is possible in two different ways.

1. *Solution of a Set of Leibnitz Equations*

The growth of the number of atoms (4) as a function of irradiation time is described by the following differential equation:

$$\frac{dN_4}{dt_b} = F_3\lambda_3 N_3 - \lambda_4 N_4 \tag{5.58}$$

where N_3 is given by equation (5.50) or (5.51), if $\lambda_2 \gg \lambda_3$. Substitution of (5.51) in (5.58) yields a linear differential equation of the first order (type equation (5.17)). Assuming $\lambda_2 \gg \lambda_3$, $\lambda_2 \gg \lambda_4$ and $N_2^0 = N_3^0 = N_4^0 = 0$, one obtains after integration between 0 and t_b:

$$N_4(t_b) = \exp(-\lambda_4 t_b)\varphi N_1^0(F_2\sigma_1 + \sigma_1')F_3 \times$$

$$\int_0^{t_b} [1 - \exp(-\lambda_3 t_b)] \exp(\lambda_4 t_b)\, dt_b$$

$$N_4(t_b) = \frac{F_3\varphi N_1^0(F_2\sigma_1 + \sigma_1')}{\lambda_4(\lambda_4 - \lambda_3)} \{\lambda_4[1 - \exp(-\lambda_3 t_b)]$$

$$- \lambda_3[1 - \exp(-\lambda_4 t_b)]\}$$

$$\boxed{\begin{aligned} D_4(t_b) = \frac{F_3\varphi N_1^0(F_2\sigma_1 + \sigma_1')}{\lambda_4 - \lambda_3} \{\lambda_4[1 - \exp(-\lambda_3 t_b)] \\ - \lambda_3[1 - \exp(-\lambda_4 t_b)]\} \end{aligned}} \tag{5.59}$$

The above equation equals equation (5.44). However the formation of ^{77}Ge by way of its isomeric form is taken into account by addition of the two reaction cross sections $F_2\sigma_1$ and σ_1'.

2. *Solution by the Bateman–Rubinson Equation*

The method of Rubinson can also be applied (see section III, C, 2), considering two separate chains:

$$(1) \xrightarrow{\sigma_1'} (3) \xrightarrow{F_3\lambda_3} (4) \xrightarrow{\lambda_4} \text{ and } (1) \xrightarrow{\sigma_1} (2) \xrightarrow{F_2\lambda_2} (3) \xrightarrow{F_3\lambda_3} (4) \xrightarrow{\lambda_4}$$

The solution for the first chain is given by equation (5.44); in the above case, this equation must be written as follows:

$$N_4'(t_b) = \frac{F_3\varphi\sigma_1' N_1^0}{\lambda_4(\lambda_4 - \lambda_3)} \{\lambda_4[1 - \exp(-\lambda_3 t_b)] - \lambda_3[1 - \exp(-\lambda_4 t_b)]\}$$

By application of equations (5.38) and (5.39) to the second chain, one finds:

$$\Lambda_1 = \varphi(\sigma_1 + \sigma_1')$$

$$\Lambda_1^* = \varphi\sigma_1$$

$$\Lambda_2 = \lambda_2; \quad \Lambda_2^* = F_2\lambda_2$$

$$\Lambda_3 = \lambda_3; \quad \Lambda_3^* = F_3\lambda_3$$

$$\Lambda_4 = \lambda_4$$

$$C_1 = \frac{1}{(\Lambda_2 - \Lambda_1)(\Lambda_3 - \Lambda_1)(\Lambda_4 - \Lambda_1)}$$

$$= \frac{1}{[\lambda_2 - \varphi(\sigma_1 + \sigma_1')][\lambda_3 - \varphi(\sigma_1 + \sigma_1')][\lambda_4 - \varphi(\sigma_1 + \sigma_1')]}$$

$$C_2 = \frac{1}{(\Lambda_1 - \Lambda_2)(\Lambda_3 - \Lambda_2)(\Lambda_4 - \Lambda_2)}$$

$$= \frac{1}{[\varphi(\sigma_1 + \sigma_1') - \lambda_2](\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)}$$

$$C_3 = \frac{1}{(\Lambda_1 - \Lambda_3)(\Lambda_2 - \Lambda_3)(\Lambda_4 - \Lambda_3)}$$

$$= \frac{1}{[\varphi(\sigma_1 + \sigma_1') - \lambda_3](\lambda_2 - \lambda_3)(\lambda_4 - \lambda_3)}$$

$$C_4 = \frac{1}{(\Lambda_1 - \Lambda_4)(\Lambda_2 - \Lambda_4)(\Lambda_3 - \Lambda_4)}$$

$$= \frac{1}{[\varphi(\sigma_1 + \sigma_1') - \lambda_4](\lambda_2 - \lambda_4)(\lambda_3 - \lambda_4)}$$

Solution of the second chain:

$$N_4''(t_b) = N_1^0\Lambda_1^*\Lambda_2^*\Lambda_3^* \sum_{i=0}^{4} C_i \exp(-\Lambda_i t)$$

$$N''(t_b) = N_1^0\varphi\sigma_1F_2\lambda_2F_3\lambda_3 \times$$

$$\left\{\frac{\exp[-\varphi(\sigma_1 + \sigma_1')t_b]}{[\lambda_2 - \varphi(\sigma_1 + \sigma_1')][\lambda_3 - \varphi(\sigma_1' + \sigma_1)][\lambda_4 - \varphi(\sigma_1 + \sigma_1')]}\right.$$

$$+ \frac{\exp(-\lambda_2 t_b)}{[\varphi(\sigma_1 + \sigma_1') - \lambda_2](\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)}$$

$$+ \frac{\exp(-\lambda_3 t_b)}{[\varphi(\sigma_1 + \sigma_1') - \lambda_3](\lambda_2 - \lambda_3)(\lambda_4 - \lambda_3)}$$

$$\left.+ \frac{\exp(-\lambda_4 t_b)}{[\varphi(\sigma_1 + \sigma_1') - \lambda_4](\lambda_2 - \lambda_4)(\lambda_3 - \lambda_4)}\right\}$$

In this example $\lambda_2 \gg \lambda_3$, $\lambda_2 \gg \lambda_4$, and if burn-up is negligible, $\varphi(\sigma_1 + \sigma_1') \ll \lambda_2, \lambda_3, \lambda_4$. Hence, the above equation can be simplified:

$$N''(t_b) \approx N_1^0\varphi\sigma_1F_2\lambda_2F_3\lambda_3\left[\frac{1}{\lambda_2\lambda_3\lambda_4} - \frac{\exp(-\lambda_3 t_b)}{\lambda_2^3}\right.$$

$$\left.+ \frac{\exp(-\lambda_3 t_b)}{\lambda_2\lambda_3(\lambda_3 - \lambda_4)} + \frac{\exp(-\lambda_4 t_b)}{\lambda_2\lambda_4(\lambda_4 - \lambda_3)}\right]$$

$$N''(t_b) \approx N_1^0\varphi\sigma_1F_2F_3\left[\frac{1}{\lambda_4} - \frac{\exp(-\lambda_3 t_b)}{\lambda_4 - \lambda_3} + \frac{\lambda_3\exp(-\lambda_4 t_b)}{\lambda_4(\lambda_4 - \lambda_3)}\right]$$

$$N_4''(t_b) \approx \frac{F_2F_3N_1^0\varphi\sigma_1}{\lambda_4(\lambda_4 - \lambda_3)}[\lambda_4 - \lambda_3 - \lambda_4\exp(-\lambda_3 t_b) + \lambda_3\exp(-\lambda_4 t_b)]$$

$$N_4''(t_b) \approx \frac{F_2F_3N_1^0\varphi\sigma_1}{\lambda_4(\lambda_4 - \lambda_3)}\{\lambda_4[1 - \exp(-\lambda_3 t_b)] - \lambda_3[1 - \exp(-\lambda_4 t_b)]\}$$

The sum of the nuclides (4) by the two paths equals $N_4'(t_b) + N_4''(t_b)$, or

$$N_4(t_b) = \frac{F_3N_1^0\varphi(F_2\sigma_1 + \sigma_1')}{\lambda_4(\lambda_4 - \lambda_3)}\{\lambda_4[1 - \exp(-\lambda_3 t_b)]$$

$$- \lambda_3[1 - \exp(-\lambda_4 t_b)]\}$$

This is identical with equation (5.59) found by the first calculation method.

As already stated, there is no essential difference between equations (5.44) and (5.59), if $\lambda_2 \gg \lambda_3$ and $\lambda_2 \gg \lambda_4$, that means: it does not matter, whether the nuclides (4) (scheme (5.56)) are formed by way of the first chain or by way of the second chain, on condition that $\sigma = F_2\sigma_1 + \sigma_1'$, i.e. the effective reaction cross section is used.

This simplification reduces the radioactive decay to the simple parent–daughter relationship (3) $\xrightarrow{\lambda_3}$ (4) $\xrightarrow{\lambda_4}$, described by equation (5.19). This equation must in this case be written as follows:

$$N_4(t_b, t) = \frac{\lambda_3}{\lambda_4 - \lambda_3} N_3(t_b)\,[\exp(-\lambda_3 t) - \exp(-\lambda_4 t)] \\ + N_4(t_b)\exp(-\lambda_4 t)$$

and this enables one to calculate the number of atoms (4) as a function of irradiation time t_b and of waiting time t. Substitution of (5.51) and (5.44), which must be written here as

$$N_4(t_b) = \frac{\varphi N_1^0(\sigma_1 + \sigma_1')}{\lambda_4(\lambda_4 - \lambda_3)}\,\{\lambda_4[1 - \exp(-\lambda_3 t_b)] - \lambda_3[1 - \exp(-\lambda_4 t_b)]\}$$

into the above equation gives (assuming that $F_2 = F_3 = 1$):

$$\begin{aligned} N_4(t_b, t) = {} & \frac{\varphi N_1^0(\sigma_1 + \sigma_1')}{\lambda_4 - \lambda_3}\,[1 - \exp(-\lambda_3 t_b)]\,[\exp(-\lambda_3 t) - \exp(-\lambda_4 t)] \\ & + \frac{\varphi N_1^0(\sigma_1 + \sigma_1')}{\lambda_4(\lambda_4 - \lambda_3)}\,\{\lambda_4[1 - \exp(-\lambda_3 t_b)] \\ & - \lambda_3[1 - \exp(-\lambda_4 t_b)]\}\exp(-\lambda_4 t) \end{aligned}$$

$$\begin{aligned} N_4(t_b, t) = {} & \frac{\varphi N_1^0(\sigma_1 + \sigma_1')}{\lambda_4(\lambda_4 - \lambda_3)}\,\{\lambda_4[1 - \exp(-\lambda_3 t_b)]\exp(-\lambda_3 t) \\ & - \lambda_4[1 - \exp(-\lambda_3 t_b)]\exp(-\lambda_4 t) \\ & + \lambda_4[1 - \exp(-\lambda_3 t_b)]\exp(-\lambda_4 t) \\ & - \lambda_3[1 - \exp(-\lambda_4 t_b)]\exp(-\lambda_4 t)\} \end{aligned}$$

Thus

$$\boxed{\begin{aligned} D_4(t_b, t) = {} & \frac{\varphi N_1^0(\sigma_1 + \sigma_1')}{\lambda_4 - \lambda_3}\,\{\lambda_4[1 - \exp(-\lambda_3 t_b)]\exp(-\lambda_3 t) \\ & - \lambda_3[1 - \exp(-\lambda_4 t_b)]\exp(-\lambda_4 t)\} \end{aligned}} \tag{5.60}$$

This has the same form as equation (5.45), σ being replaced by $\sigma_1 + \sigma_1'$.

(E) "SECOND ORDER" REACTIONS

When neutron activation analysis is used to determine trace elements at very low levels (ppm, ppb . . .), high sensitivity techniques are necessary. Sensitivity can be enhanced by using large reactor fluxes and long irradiation times. But under these conditions neutron induced second order nuclear reactions can lead to significant interference in determining a trace element as additional amounts of the measured radionuclide can be produced (7).

This second order interference is a systematic error, produced by successive reactions of the type:

$${}^{A}_{Z}E\ (\mathrm{n}, \gamma)\ {}^{A+1}_{Z}E \left\{ \begin{array}{l} \xrightarrow{\beta^-} {}^{A+1}_{Z+1}E\ (\mathrm{n}, \gamma)\ {}^{A+2}_{Z+1}E \\ \xrightarrow{\beta^+/EC} {}^{A+1}_{Z-1}E\ (\mathrm{n}, \gamma)\ {}^{A+2}_{Z-1}E \end{array} \right. \tag{5.61}$$

or, schematically

$$\begin{tikzcd} (1) \arrow[r, "\mathrm{n},\gamma", "\sigma_1 \atop \lambda_1 = 0"'] & (2) \arrow[r, "\beta^- : F_2\lambda_2"] \arrow[d, "\mathrm{n},\gamma"', "\sigma_2"] \arrow[dr, "\beta^+/EC : F_2'\lambda_2"] & (3) \arrow[r, "\mathrm{n},\gamma", "\sigma_3 \atop \lambda_3 = 0"'] & (4) \arrow[r, "\beta^-(\beta^+/EC)"] \arrow[d, "\mathrm{n},\gamma"', "\sigma_4"] & (5)\ \text{(stable)} \\ {} & (3') & & {} & \end{tikzcd} \tag{5.62}$$

where F_2 is the fraction of disintegrations of nuclides (2) producing nuclides (3). In most cases $F_2 = 1$ and $F_2' = 0$ (or $F_2 = 0$ and $F_2' = 1$), furthermore $\sigma_2 = 0$, $\sigma_4 = 0$ or are negligible: $\varphi\sigma_2 \ll \lambda_2$, $\varphi\sigma_4 \ll \lambda_4$ (if the flux is not too high and burn-up negligible). However calculations will be given for the general case.

Example: Second order reaction on a silicon matrix:

$${}^{30}\mathrm{Si}\ (\mathrm{n}, \gamma)\ {}^{31}\mathrm{Si} \xrightarrow{\beta^-} {}^{31}\mathrm{P}\ (\mathrm{n}, \gamma)\ {}^{32}\mathrm{P} \xrightarrow{\beta^-}$$

can give erroneous results in determining the phosphorus content by neutron activation analysis, because the same nuclide (^{32}P) is formed as in the reaction

$${}^{31}\mathrm{P}\ (\mathrm{n}, \gamma)\ {}^{32}\mathrm{P} \xrightarrow{\beta^-}$$

Other examples:

Iridium in osmium: $^{190}\mathrm{Os}\ (\mathrm{n}, \gamma)\ {}^{191}\mathrm{Os} \xrightarrow{\beta^-} {}^{191}\mathrm{Ir}\ (\mathrm{n}, \gamma)\ {}^{192}\mathrm{Ir} \xrightarrow{\beta^-}$

$${}^{191}\mathrm{Ir}\ (\mathrm{n}, \gamma)\ {}^{192}\mathrm{Ir} \xrightarrow{\beta^-} (^{192}\mathrm{Ir}\ \text{measured})\ (8)$$

Thallium in mercury: $^{202}\mathrm{Hg}\,(\mathrm{n}, \gamma)\,^{203}\mathrm{Hg} \xrightarrow{\beta^-} {}^{203}\mathrm{Tl}\,(\mathrm{n}, \gamma)\,^{204}\mathrm{Tl} \longrightarrow$
$^{203}\mathrm{Tl}\,(\mathrm{n}, \gamma)\,^{204}\mathrm{Tl} \longrightarrow$ (^{204}Tl measured).

The rates of nuclide transformation in scheme (5.62) are described by the following system of differential equations:

$$\begin{aligned}
\frac{\mathrm{d}N_1}{\mathrm{d}t_b} &= -N_1\varphi\sigma_1 \\
\frac{\mathrm{d}N_2}{\mathrm{d}t_b} &= N_1\varphi\sigma_1 - \lambda_2 N_2 - N_2\varphi\sigma_2 \\
\frac{\mathrm{d}N_3}{\mathrm{d}t_b} &= F_2\lambda_2 N_2 - N_3\varphi\sigma_3 \\
\frac{\mathrm{d}N_4}{\mathrm{d}t_b} &= N_3\varphi\sigma_3 - \lambda_4 N_4 - N_4\varphi\sigma_4
\end{aligned} \tag{5.63}$$

These equations can be solved in the classical way, but the solution for N_3 is laborious, since it is a much more complicated function than N_1. The next solution, for N_4, is even more tedious. So it is more simple to use directly the Bateman–Rubinson solution.

$$\begin{aligned}
\Lambda_1 &= \Lambda_1^* = \varphi\sigma_1 \\
\Lambda_2 &= \lambda_2 + \varphi\sigma_2 \\
\Lambda_2^* &= F_2\lambda_2 \\
\Lambda_3 &= \Lambda_3^* = \varphi\sigma_3 \\
\Lambda_4 &= \lambda_4 + \varphi\sigma_4
\end{aligned}$$

Substituting these values in equation (5.38), gives

$$N_4 = N_1^0\varphi^2\sigma_1\sigma_3F_2\lambda_2\sum_{i=1}^{i=4} C_i \exp\,(-\Lambda_i t_b) \tag{5.64}$$

where

$$\begin{aligned}
C_1 &= \frac{1}{(\Lambda_2 - \Lambda_1)\,(\Lambda_3 - \Lambda_1)\,(\Lambda_4 - \Lambda_1)} \\
&= \frac{1}{(\lambda_2 + \varphi\sigma_2 - \varphi\sigma_1)\,(\varphi\sigma_3 - \varphi\sigma_1)\,(\lambda_4 + \varphi\sigma_4 - \varphi\sigma_1)}
\end{aligned}$$

$$\begin{aligned}
C_2 &= \frac{1}{(\Lambda_1 - \Lambda_2)\,(\Lambda_3 - \Lambda_2)\,(\Lambda_4 - \Lambda_2)} \\
&= \frac{1}{(\varphi\sigma_1 - \lambda_2 - \varphi\sigma_2)\,(\varphi\sigma_3 - \lambda_2 - \varphi\sigma_2)\,(\lambda_4 + \varphi\sigma_4 - \lambda_2 - \varphi\sigma_2)} \\
C_3 &= \frac{1}{(\Lambda_1 - \Lambda_3)\,(\Lambda_2 - \Lambda_3)\,(\Lambda_4 - \Lambda_3)} \\
&= \frac{1}{(\varphi\sigma_1 - \varphi\sigma_3)\,(\lambda_2 + \varphi\sigma_2 - \varphi\sigma_3)\,(\lambda_4 + \varphi\sigma_4 - \varphi\sigma_3)} \\
C_4 &= \frac{1}{(\Lambda_1 - \Lambda_4)\,(\Lambda_2 - \Lambda_4)\,(\Lambda_3 - \Lambda_4)} \\
&= \frac{1}{(\varphi\sigma_1 - \lambda_4 - \varphi\sigma_4)\,(\lambda_2 + \varphi\sigma_2 - \lambda_4 - \varphi\sigma_4)\,(\varphi\sigma_3 - \lambda_4 - \varphi\sigma_4)}
\end{aligned}$$

The Bateman–Rubinson equation is tedious to solve by hand and often leads to loss in significance in performing the summations of the exponential terms $C_i \exp\,(-\Lambda_i t_b)$. For this reason the aid of a computer with double precision is highly desirable. A more detailed discussion about second order interference as a source of error in activation analysis is given in Chapter 10, section II, C, 3.

References

1. Nervik, W. E., *Applications of Computers to Nuclear and Radiochemistry.* Washington, NAS–NS 3107, 1963, p. 9.
2. Cumming, J. B., *ibid.*, p. 25.
3. Gardner, D. G., and Gardner, J. C., *ibid.*, p. 33.
4. Gardner, D. G., Gardner, J. C., Lausch, G., and Meinke, W., *J. Chem. Phys.*, **31**, 178 (1959).
5. Bateman, H., *Proc. Cambridge Phil. Soc.*, **15**, 423 (1910).
6. Rubinson, W., *J. Chem. Phys.*, **17**, 542 (1949).
7. Ricci, E., and Dyer, F. F., *Nucleonics*, **22**(6), 45 (1964).
8. Gijbels, R., and Hoste, J., *Proc. Conf. Modern Trends in Activation Analysis*, College Station, Texas U.S.A., April 19–22, 1965, p. 129.