Specific Activity after Intermittent Irradiation

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To SIMPLIFY CALCULATION of the final specific activity obtained when samples are activated by intermittent irradiation in a research reactor, we have developed a graphical method that uses "normalized" growth and decay curves. These curves coupled with a sheet of transparent graph paper and a bit of arithmetic are all the tools one needs. If flux is constant for all irradiation periods (and constant throughout each irradiation period) the theory allows a direct graphical solution, but when irradiations are made at several flux levels, one must find from the graph a component of the total activity for each irradiation and then sum the components to get the total normalized specific activity.

Theory

Specific activity in curies per gram is given by the formula

$$A_\infty = 0.163\sigma\phi a/M \qquad (1)$$

where σ is activation cross section in barns, ϕ is neutron flux in multiples of 10^{12} n/cm²/sec, a is isotopic abundance in per cent and M is atomic weight.

To develop a generalized case one can normalize activities and times:

normalized activity $= \alpha = A/A_\infty$
normalized time $= \theta = t/T_{1/2}$

where A is activity, A_∞ is saturation activity, $T_{1/2}$ is half-life and t is any irradiation or cooling time. From these factors come the normalized growth curve $G = \ln \alpha(\theta_i)$ and normalized decay curve $D = \ln \alpha(\theta_c)$ (see figure).

If flux is constant the normalized activity after a sequence of irradiation for a period θ_{i1}, cooling for a period θ_{c1} and irradiation for a period θ_{i2} will be given by:

$$\alpha_{(\theta_{i1}+\theta_{c1}+\theta_{i2})} = (1 - e^{-0.693\theta_{i1}})e^{-0.693(\theta_{c1}+\theta_{i2})} + (1 - e^{-0.693\theta_{i2}}) \qquad (2)$$

This equation can be transformed into

$$\alpha_{(\theta_{i1}+\theta_{c1}+\theta_{i2})} = 1 - e^{-0.693\theta_{i2}}[1 - (1 - e^{-0.693\theta_{i1}})e^{-0.693\theta_{c1}}] \qquad (3)$$

The second term in the brackets in Eq. 3 equals the normalized activity $\alpha(\theta_{i1} + \theta_{c1})$ obtained before the second irradiation. An equivalent amount of normalized activity could be obtained after irradiation for a period θ_{ix} without cooling

$$\alpha_{(\theta_{ix})} = \alpha_{(\theta_{i1}+\theta_{c1})} = 1 - e^{-0.693\theta_{ix}} \qquad (4)$$

If this quantity replaces the second term in the brackets of Eq. 3 one gets

$$\alpha_{(\theta_{i1}+\theta_{c1}+\theta_{i2})} = 1 - e^{-0.693\theta_{i2}}[1 - (1 \; e^{-0.693\theta_{ix}})] = 1 - e^{0.693(\theta_{ix}+\theta_{i2})} = \alpha_{(\theta_{ix}+\theta_{i2})} \qquad (5)$$

Thus the normalized activity after the sequence $\theta_{i1} + \theta_{c1} + \theta_{i2}$ is equal to the normalized activity obtained from an irradiation period θ_{i2} that follows an irradiation period θ_{ix} chosen to give the equivalent of the first irradiation and cooling. This equivalent, and in fact the whole solution, can be best obtained graphically in a manner that we shall presently explain.

Flux factors. If the flux ϕ is not the same for all irradiation periods θ_{i1}, θ_{i2}, . . . but is constant throughout each irradiation, the general equation is

$$A = 0.163\sigma \frac{a}{M} [\phi_1(1 - e^{-0.693\theta_{i1}}) e^{-0.693(\theta_{c1}+\theta_{i2}+\theta_{c2}+\cdots)} + \phi_2(1 - e^{-0.693\theta_{i2}}) e^{-0.693(\theta_{c2}+\theta_{i3}+\cdots)} + \cdots] \qquad (6)$$

The fluxes ϕ_1, ϕ_2, . . . are now expressed as multiples of one of them, for example ϕ_1, so that Eq. 6 becomes:

$$A = 0.163\sigma\phi \frac{a}{M} [(1 - e^{-0.693\theta_{i1}})e^{-0.693\theta_{c1}+\theta_{i2}+\theta_{c2}+\cdots} + \frac{\phi_2}{\phi_1}(1 - e^{-0.693\theta_{i2}})e^{-0.693(\theta_{c2}+\theta_{i3}+\cdots)} + \cdots] \qquad (7)$$

The factor ϕ_x/ϕ_1 is equivalent to a period $\theta(\phi_x)$ on the decay curve D given by

$$\frac{\phi_x}{\phi_1} = e^{-0.693\theta(\phi_x)} \qquad (8)$$

To account for this factor, the point obtained on the G curve for $(1 - e^{-0.693\theta_{1x}})$ is displaced on the D curve $\theta(\phi_x)$ units to the left or to the right, depending on whether ϕ_x/ϕ_1 is larger or smaller than one. The flux factors in Eq. 7 are thus converted to a factor to be added (or subtracted) to the decay factors, and the expression for normalized activity becomes

$$\alpha = (1 - e^{-0.693\theta_{i1}}) e^{-0.693(\theta_{c1}+\theta_{i2}+\theta_{c2}+\cdots)} + (1 - e^{-0.693\theta_{i2}})e^{-0.693(\theta_{\phi2}+\theta_{c2}+\theta_{i3}+\cdots)} + (1 - e^{-0.693\theta_{i3}})e^{-0.693(\theta_{\phi3}+\theta_{c3}+\cdots)} + \cdots \qquad (9)$$

The different terms of Eq. 9 must be obtained separately from the graph and then added.

How to Apply the Theory

Consider two representative problems:

I At a constant flux of 10^{13} n/cm²/sec irradiate a sodium sample for 4 hr 30 min, allow it to cool for 6 hr, irradiate at the same flux for 4 hr 12 min and cool 6 hr. Find specific activity.

II When second irradiation is done

at a different flux, 1.5×10^{13} n/cm²/sec, (no other conditions changed) what is final specific activity? (For Na^{24}, $T_{1/2} = 15$ hr; for Na^{23} $\sigma = 0.56$ barns, $a = 100\%$).

The solution to Problem I (constant flux) is almost wholly graphical and is obtained as follows:

Graphical Procedures (constant ϕ)

1. Express irradiation periods as $\theta_{i1}, \theta_{i2}, \ldots$ and cooling periods as θ_{c1}, θ_{c2}, (in both problems $\theta_{i1} = 0.30$, $\theta_{c1} = 0.40$, $\theta_{i2} = 0.28$ and $\theta_{c2} = 0.40$).

2. Superimpose a sheet of transparent graph paper on the chart and draw coordinates.

3. Mark on the transport sheet the ordinate of the G curve for θ_{i1} (mark x_1).

4. Move the transparent sheet vertically or horizontally until mark x_1 coincides with D curve.

5. Mark the ordinate of the D curve for $\theta_{i1} + \theta_{c1}$ (mark x_2).

6. Adjust the origins of the two sheets to coincide, then move the transparent sheet horizontally, until mark x_2 coincides with G curve. This produces the operation shown in Eq. 4; the abscissa corresponding to x_2 is θ_{ix}.

7. Mark the ordinate of the G curve for $\theta_{ix} + \theta_{i2}$ (mark x_3).

8. Repeat step 4.

9. Mark the ordinate of the D curve for $\theta_{ix} + \theta_{i2} + \theta_{c2}$ (mark x_4).

10. Do likewise for all irradiation periods θ_i and cooling periods θ_c.

11. Read on the ordinate the normalized activity for the last mark made. For problem I this value is $\alpha = 0.22$.

12. Calculate A_∞ from the known flux, cross-section, abundance and atomic weight, and multiply by the normalized activity to get the desired specific activity. So, $A_\infty = 0.163\sigma\phi\, a/M = 0.163 \times 0.56 \times 10 \times {}^{100}\!/_{23} = 3.99$ c/gm and $A = 0.22A_\infty = 0.881$ c/gm.

With several flux values (Problem II) terms that make up the normalized specific activity in the equation must be obtained separately and then summed. Use the following procedure:

A. Compute the flux factors. In Problem II this is $\phi_2/\phi_1 = 1.5$.

B. Determine the $\theta_{\phi 2}$ (on the θ axis) that is equivalent to the flux factor on the ϕ_x/ϕ_1 scale. In this problem the length involved is from 1 to 1.5 on the ϕ_x/ϕ_1 axis and $\theta_{\phi 2}$ equals -0.585 (minus since ratio is >1).

C. Substitute values in Eq. 9. The normalized activity becomes

$$\alpha = (1 - e^{-0.693\times 0.30})e^{-0.693(0.40+0.28+0.40)} + (1 - e^{-0.693\times 0.28})e^{-0.693(0.40-0.585)}$$

D. Find the value of the first term by locating $\theta_{i1}(0.30)$ on the G curve and correcting for all subsequent decay $(1.08 = \theta_{c1} + \theta_{i2} + \theta_{c2})$ on the D curve. Read the value on the ordinate—it is 0.089.

E. Find the value of the second term by locating θ_{i2} on the G curve and correcting for both flux factor and decay $(-0.185 = \theta_{\phi 2} + \theta_{c2})$ along the D curve. Again read the α value on the ordinate (0.200).

F. Follow the same procedure with additional irradiations.

G. Sum the terms (0.289).

H. Compute specific activity from total normalized activity. For the second problem this is $0.289 \times 3.97 = 1.15$ c/gm.

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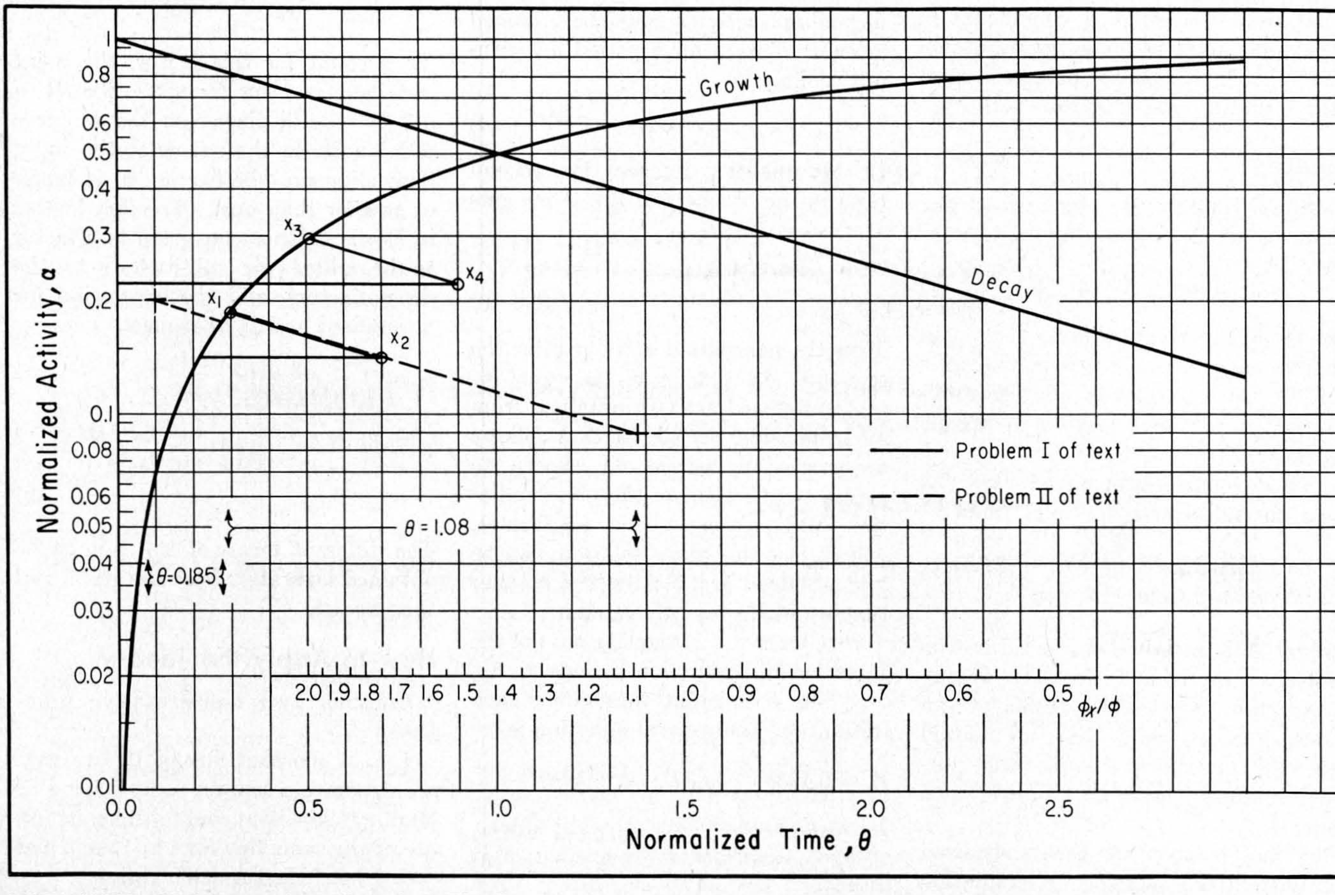

NORMALIZED GROWTH AND DECAY CURVES offer a quick method for calculating induced activity in neutron-irradiated samples