

Chapter 5

Laws of radioactive decay

In this Chapter we will deal with the analytics of coupled and decoupled radioactive decay. While these analytic results are important touchstones, the numerical solution of the coupled differential equations is a more direct approach. The direct approach is presented in a MATHCAD template. However before exercising that template, one should digest the analytics presented in this chapter.

5.1 Fundamentals of decay of a species

An isolated quantum state undergoes 1st order decay kinetics. Radioactive decay (decay of a collection of unstable nuclei) is the prototypical example of this kinetics. The decay rate is proportional to the number of nuclei in the parent population N , with the proportionality constant λ (time⁻¹) called the **decay constant**.

$$\begin{aligned} \frac{\text{Decays}}{\text{Unit time}} &\propto \text{Number of nuclei available.} \\ -\frac{dN}{dt} &= \lambda \cdot N. \end{aligned}$$

Rearranging the above and integrating we find

$$\begin{aligned} dN &= -\lambda N dt, \\ \int_{N_0}^N \frac{dN'}{N'} &= \int_{t'=0}^t -\lambda dt, \\ \ln N]_0^t &= -\lambda t']_0^t, \\ \ln N(t) - \ln N(0) &= -\lambda t, \\ \ln \frac{N(t)}{N(0)} &= -\lambda t, \\ N(t) &= N(0) \cdot e^{-\lambda t}. \end{aligned} \quad (5.1)$$

The **half live** $t_{1/2}$ is the time at which half of the initial number of atoms survive, while the **mean time** or average life is the time at which the undecayed fraction is $1/e=1/2.718 = 0.367$. Note that $\tau > t_{1/2}$ (more decay

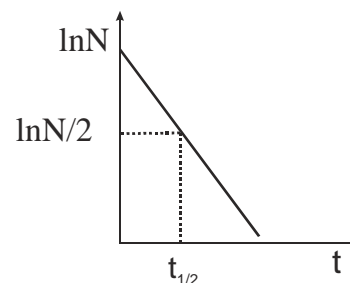


Figure 5.1: Exponential decay. The $t_{1/2}$ is indicated.

in a time τ than in $t_{1/2}$.) The mean time is the reciprocal of the decay constant $\tau = \frac{1}{\lambda}$, while $t_{1/2} = \ln 2/\lambda = 0.693/\lambda = 0.693 \cdot \tau$, thus $\tau = 1.4427 t_{1/2}$.

The reciprocal relationship between τ and λ makes these quantities appealing to use and can be appreciated by the following integration.

$$\begin{aligned} \tau &= \frac{\int_0^\infty t \cdot dN}{\int_0^\infty dN} = -\frac{1}{N(0)} \int_0^\infty (-) t \lambda N dt \\ &= \frac{1}{N(0)} \int_0^\infty t \lambda N(0) e^{-\lambda t} dt = \lambda \int_0^\infty t^1 e^{-\lambda t} dt \\ &= \lambda \frac{1!}{\lambda^2} = \frac{1}{\lambda} \text{ (after integration by parts).} \end{aligned}$$

The number of decays per unit time or the activity A (decays/s), of a given species, is $\lambda_i \cdot N_i(t)$ and the count rate CR (counts/s) is the activity times a detection efficiency ϵ_i for the particular radiation.

$$\begin{aligned} A_i(t) &= \lambda_i \cdot N_i(t) = A_i(0) e^{-\lambda t}, \\ CR_i(t) &= \epsilon_i \cdot A_i(t) \end{aligned}$$

5.2 Independently decaying activities

In a mixture of independently decaying activities

$$A_{tot}(t) = \sum_{i=1}^n A_i = \sum_{i=1}^n \lambda_i [N_i(t)].$$

$$A_{tot}(t) = \sum_{i=1}^n A_i(0)e^{-\lambda_i t} = \sum_{i=1}^n \lambda_i [N_i(0)e^{-\lambda_i t}].$$

If the half lives are sufficiently different, then they can be resolved from one another by peeling off the longest half live component first, the next one, etc. (Of course one can also just do a least squares fit.) On the other hand, if the half lives are similar numerically, it is very difficult to resolve them.

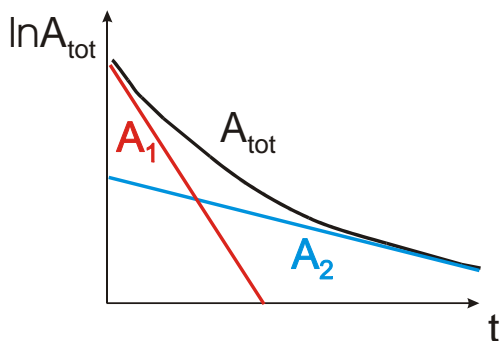


Figure 5.2: Resolving graphically two activities.

If one has two components, and the half lives are known, then the following trick, shown in Fig. 5.3, can be used to extract the initial activities $A_i(0)$.

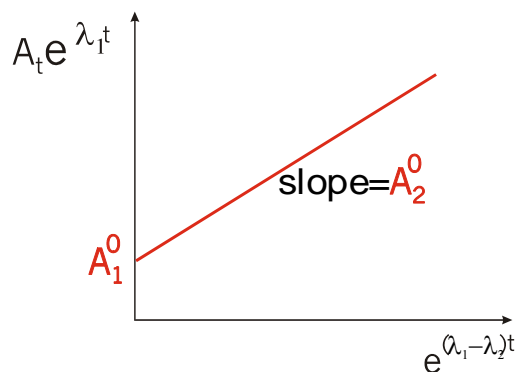


Figure 5.3: Trick to resolve two components with similar but known half lives.

$$A_{tot} = A_1^0 e^{-\lambda_1 t} + A_2^0 e^{-\lambda_2 t}, \text{ multiply by } e^{\lambda_1 t},$$

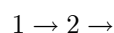
$$A_{tot} e^{\lambda_1 t} = A_1^0 + A_2^0 e^{(\lambda_1 - \lambda_2)t}, \text{ which is of the form}$$

$$y(x) = a + b \cdot x.$$

5.3 Growth and Decay

5.3.1 Master equations

Consider two genetically related activities, that is species 1 (*the parent*) decays into species 2 (*the daughter*) which is also decaying.



The amount of the parent is determined by its decay rate

$$\frac{dN_1}{dt} \equiv \dot{N}_1 = -\lambda_1 N_1(t), \quad (5.2)$$

The rate of net growth of the daughter (2) equals the growth rate from the decay of the parent minus its own decay rate

$$\frac{dN_2(t)}{dt} \equiv \dot{N}_2 = \lambda_1 N_1(t) - \lambda_2 N_2(t) \quad (5.3)$$

The two differential equations Eqs. 5.2 and 5.3 are sometimes called **master equations**. These equations plus the initial conditions $N_1(0)$ and $N_2(0)$ can always be solved numerically (see provided template.) This brute force method is useful in complicated cases involving branching and feeding. However in the case we have constructed (single parent and daughter) the set of coupled DEQ's has an analytic solution.

5.3.2 Analytic solution

Dropping the indication of a time dependence, one can rewrite the master equation for the daughter as

$$\dot{N}_2 + \lambda_2 N_2 - \lambda_1 N_1 = 0$$

This is a linear 1st order differential equation with a general solution

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t}. \quad (5.4)$$

This solution blows up in the special case $\lambda_2 = \lambda_1 \equiv \lambda$. In this unusual case there is another analytic solution

$$N_2 = N_1^0 \lambda t e^{-\lambda t}.$$

If we multiply N_2 by λ_2 we get

$$A_2 = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + A_2^0 e^{-\lambda_2 t}. \quad (5.5)$$

The first terms in Eqs. 5.4 and 5.5 represents the decay of the daughters generated from the decay of the parent while the second treats the decay of any daughter present initially.

5.3.3 Cases

The following misnamed special cases are useful benchmarks. (I say misnamed because there is never an equilibrium in the thermodynamic sense.)

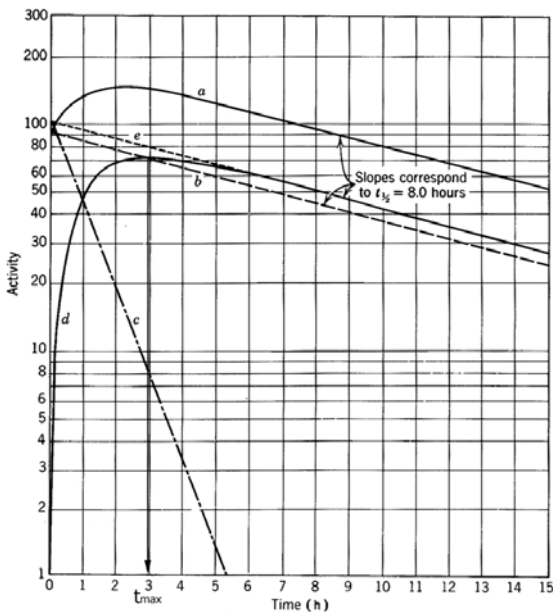


Figure 5.4: Transient equilibrium: (a) total activity of an initially pure parent fraction; (b) activity due to parent ($t_{1/2} = 8.0$ hr); (c) decay of freshly isolated daughter fraction ($t_{1/2} = 0.80$ hr); (d) daughter activity growing in freshly purified parent fraction; (e) total daughter activity in parent-plus-daughter fractions. (from FKMM, p. 195).

Transient “Equilibrium”

If the daughter decay rate is larger than the parent $\lambda_2 > \lambda_1$ ($t_{1/2}(2) < t_{1/2}(1)$), after some time $e^{-\lambda_2 t} \ll e^{-\lambda_1 t}$

and

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 e^{-\lambda_1 t} = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1, \text{ and} \quad (5.6)$$

$$\frac{N_2}{N_1} = \frac{\lambda_1}{\lambda_2 - \lambda_1} = \text{constant, but as}$$

$$A_2 = \lambda_2 N_2, \quad A_1 = \lambda_1 N_1 \Rightarrow$$

$$\frac{A_2}{A_1} = \frac{\lambda_2}{\lambda_2 - \lambda_1} = \text{constant.} \quad (5.7)$$

This condition, constant ratio of the parent/daughter atoms and activities, is called “**Transient Equilibrium**” and is illustrated in Fig. 5.4.

The maximum daughter decay rate, from a freshly purified parent fraction, is obtained by setting the time derivative of Eq. 5.4 equal to zero

$$t_{\max} = \frac{1}{\lambda_2 - \lambda_1} \ln \frac{\lambda_2}{\lambda_1}. \quad (5.8)$$

Secular “Equilibrium”

In the extreme case the parent does not decay appreciably for several half lives of the daughter $\lambda_2 \gg \lambda_1$, then

$$\frac{N_2}{N_1} = \frac{\lambda_1}{\lambda_2}$$

or

$$\lambda_1 N_1 = \lambda_2 N_2,$$

$$A_1 = A_2.$$

This is called “**secular equilibrium**” and is illustrated in Fig. 5.5(A). This case is found in the 3 natural radioactive decay chains. The reason for this is that for these chains to be “natural”, the parent must have a $t_{1/2}$ of the order of the age of the universe.

The number of atoms N_2 at any time t [curve (d) in Fig. 5.5(A)] is obtained from Eq. 5.4 by appreciating that if λ_1 is tiny $e^{-\lambda_1 t} \approx 1$ and λ_1 can be dropped in the sum.

$$N_2 = \frac{\lambda_1}{\lambda_2} N_1^0 (1 - e^{-\lambda_2 t}) \text{ or}$$

$$\lambda_2 N_2 = \lambda_1 N_1^0 (1 - e^{-\lambda_2 t}) \text{ and} \quad (5.9)$$

$$A_2 = A_1^0 (1 - e^{-\lambda_2 t}).$$

No Equilibrium

If the parent decays faster than the daughter $\lambda_1 > \lambda_2$, the daughter grows, reaches a maximum (see Eq. 5.8) and then (at times long compared with the parent half life) decays with its own $t_{1/2}$. An example is given in Fig. 5.5(B).

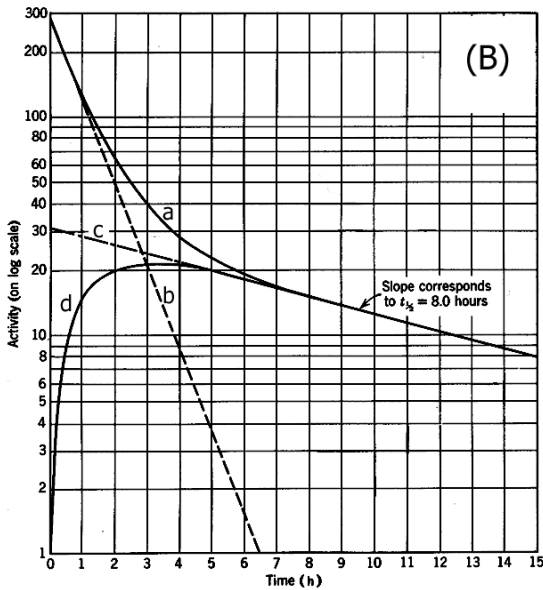
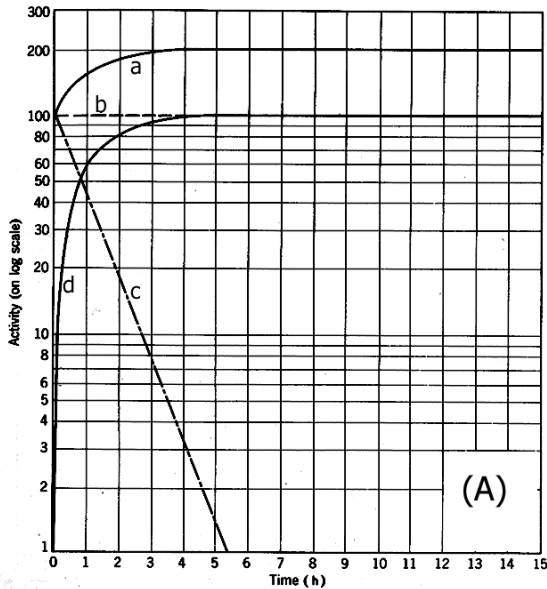


Figure 5.5: (A) **Secular equilibrium**: (a) **total activity** of an initially pure parent fraction; (b) **parent activity** ($t_{1/2} = \infty$), this is also the total activity in parent-plus-daughter fractions; (c) decay of freshly isolated daughter fraction ($t_{1/2} = 0.80$ hr); (d) **daughter activity** in a freshly purified parent fraction. (B) **No equilibrium**: (a) **total activity**; (b) **parent activity** ($t_{1/2} = 0.80$ hr); (c) extrapolation of initial decay curve to time zero; (d) **daughter activity** in initially pure parent. (From FKMM pp. 196 - 197).

Branched decay

If species 1 decays to *both* 2 and 3, as is the case for odd-odd ^{64}Cu , one must define separate rate constants for each branch, see Fig. 5.6.

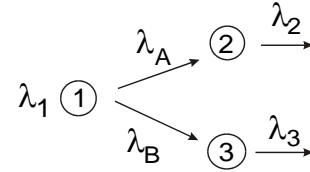


Figure 5.6: Branched decay.

Now, $\lambda_1 = \lambda_A + \lambda_B = f_A\lambda_1 + f_B\lambda_1$, where f_A and f_B are the decay fractions (or branching fractions with branching ratios of $\frac{f_i}{\sum_j f_j}$). The master equations are

$$\begin{aligned} \dot{N}_1 &= -\lambda_1 N_1 = -(\lambda_A + \lambda_B) N_1, \\ \dot{N}_2 &= \lambda_A N_1 - \lambda_2 N_2 \\ \dot{N}_3 &= \lambda_B N_1 - \lambda_3 N_3 \end{aligned}$$

Note,

$$t_{1/2}(1) = \frac{\ln 2}{\lambda_1} = \frac{\ln 2}{\lambda_A + \lambda_B}.$$

and

$$\lambda_A = f_A \lambda_1.$$

One often defines a “**partial half-life**” for species 1 for the decay path A, $t_{1/2}^A(1) = \frac{\ln 2}{\lambda_A}$. This would be the half-life of the parent IF path A were the only one available.

5.3.4 Irradiations

Now let us consider the production of an activity by some sort of irradiation. Perhaps neutron irradiation in a reactor or bombardments of charged particles from an accelerator. Lets assume we have a constant flux (or beam intensity) incident on a target. The rate of formation of the species of interest will be constant during the irradiation, but the formed species (if it is unstable) will also be decaying during the irradiation.

$$\begin{aligned} \frac{dN}{dt} &= (\text{Rate of Formation}) - (\text{Rate of Decay}) \\ &= R - \lambda N(t). \end{aligned}$$

Integrating this equation for a bombardment time t_b gives the same form as found in secular equilibrium, Eq. 5.9 with the replacements $\lambda_1 N_1^0 = R$, $\lambda_2 = \lambda$, and $t = t_b$. (The reason for the analytic similarity, is that the assumption has been made that the number of targets is

not changing in time, this the formation rate is constant. This is identical to the secular equilibrium that there is a negligible decay of the parent in the decay time.) Thus the disintegration rate of the produced activity at the end of bombardment is

$$\lambda N(t_b) = R (1 - e^{-\lambda t_b}),$$

or for the number of atoms

$$N(t_b) = \frac{R}{\lambda}(1 - e^{-\lambda t_b}).$$

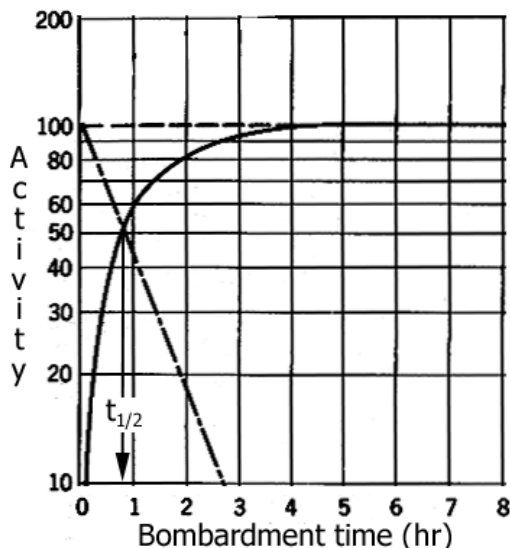


Figure 5.7: Irradiation profile for a species with $t_{1/2} = 0.80$ hr.

1. If t_b is short, then the formation rate is larger than the decay rate.
2. If t_b is much longer than $\tau = 1/\lambda$ ($e^{-\lambda t_b} \rightarrow 0$), then $N(t_b) = R/\lambda$ and a steady-state condition has been achieved.
3. Analytic solutions for the equations for the more general cases, with many members in a decay sequence, are available (see FKMM, p. 198 - 201.) However, today it is probably easier to just solve the master equations numerically (see template.)

Examples

1. Considering the case of neutron irradiation in a reactor,

$$R = N[\Phi(\frac{\#}{cm^2 s})]\{\sigma(cm^2)\}.$$

with:

- (a) N “target nuclei”
- (b) a neutron flux of $\Phi(\frac{\#}{cm^2 s})$ and
- (c) a cross section of $\sigma(cm^2)$.

2. In a beam experiment how one calculates the production rate depends on whether the target is thick or thin.

- (a) If the target is sufficiently thin that the beam goes through with little energy loss (and the production cross section does not change)

$$R_{thin} = [I'(\frac{part}{s})]\{N'(\frac{nuclei}{cm^3})t(cm)\}[\sigma'(cm^2)]$$

$$R_{thin}(s^{-1}) = 3.76 \cdot 10^6 \frac{I(na)}{q} \left\{ \frac{T(\frac{mg}{cm^2})}{A(g)} \right\} [\sigma(b)]$$

- i. $I' = 6.24 \cdot 10^9 I(na)/q$ with q being the charge state of the ion
- ii. $N' = \frac{\rho(g/cm^3)}{A(g/mole)} \cdot 6.02 \cdot 10^{23} (\frac{atoms}{mole})$
- iii. $\sigma'(cm^2) = \sigma(b) \cdot 10^{-24}$
- iv. $T(\frac{mg}{cm^2}) = \rho(\frac{g}{cm^3}) \cdot t(cm) 10^3 (\frac{mg}{g})$

- (b) If the target is thick, one must integrate over the changing cross section $\sigma(E)$. (Here we are indicating the functional dependence rather than the units.)

$$R_{thick} = \int_{init.E}^{final.E} R_{thin}(E)$$

3. In the case of a scattering beam experiment the count rate at a given angle $d\Omega(sr)$ would be

$$R(\frac{cts}{s}) = 3.78 \cdot 10^3 \left[\frac{d\sigma}{d\Omega}(\frac{mb}{sr}) \right] \left\{ \frac{I(na)}{q} \right\} \left[\frac{T(\frac{mg}{cm^2})}{A_T} \right] d\Omega(sr)$$

5.3.5 Natural Line width

The decay constant λ or the mean lifetime τ discussed thus far refer to long-lived species, where by the lifetimes are measured from a statistical averaging from a lot of decays. However, for each quantum level we can ask the question: How well do we know the energy of the level if the mean life τ is known? The answer is obtained through the uncertainty principle which can be written in terms of energy-time as well as position momentum.

$$\Delta x \cdot \Delta p \geq \frac{\hbar}{2} \text{ or}$$

$$\Delta E \cdot \tau \geq \frac{\hbar}{2}$$

where ΔE is the natural line width and τ is the mean lifetime. For example if:

1. $\tau = 1 \cdot 10^{-6} s$, $\Delta E = \frac{6.58 \times 10^{-16} eV \cdot s}{2 \cdot 10^{-6}} = 3.3 \cdot 10^{-10} eV$
2. $\tau = 1 \times 10^{-16} s$, $\Delta E = \frac{6.58 \times 10^{-16} eV \cdot s}{2 \times 10^{-16}} = 3.3 eV$.