RADIOACTIVE DECAY

Radioactive materials may either originate from natural sources or be created through technological processes. Naturally radioactive materials include carbon-14, potassium-40, and thorium and uranium isotopes and their progeny. Neutron transmutation of stable isotopes into radioisotopes is a method of artificially creating radioactive material. Of interest to the study of soft errors in circuits are the heavy decay chains of uranium and thorium. To begin our study of radioactivity, we first examine simple radioactive decay and we define terms such as half-life, decay constant, and activity.

Basic Balance Equation

The basic balance equation is a useful starting point for many analyses:

Rate of Change = Production (Inflow) – Losses (Outflow)

Simple Decay Chain

For a simple radioactive decay chain, the parent radionuclide decays to a stable product. Let $N(t)$ represent the parent radionuclide at time t, where N could be in units of total atoms (n) or atom density (N). We assume in this discussion that no production of the radionuclide occurs after $t = 0$, so there is an initial number of atoms equal to $N(0)$. These radioactive atoms then decay according the *decay constant* (λ), which is a probability per unit time that an individual atom decays, and which can be expressed in terms of the half-life ($t_{½}$) of the substance, $\lambda = \ln(2)/t_{½}$. The decay rate is a nuclear property independent of (1) temperature, (2) pressure, (3) chemical form of the isotope, and (4) physical state of the substance. Using the basic balance equation above, a first-order differential equation describing $N(t)$ is established

$$\frac{dN}{dt} = -\lambda N(t) \tag{1}$$

Laplace transforming the differential equation yields:

$$s\,N(s) - N(0) = -\lambda N(s) \tag{2}$$

The above expression is algebraically manipulated to isolate the variable of interest, $N(s)$:

$$(s+\lambda)\,N(s) = N(0)$$

$$N(s) = \frac{N(0)}{(s+\lambda)} \tag{3}$$

Finally, the inverse Laplace transform is taken to determine the time dependent concentration of the parent radionuclide for $t \geq 0$:

$$N(t) = N(0)\,e^{-\lambda t} = N(0)\left(\tfrac{1}{2}\right)^{t/t_{½}} \tag{4}$$

where the decay constant (λ) and half-life of the radionuclide ($t_{½}$) are related by

$$t_{½} = \frac{\ln(2)}{\lambda} \tag{5}$$

The average (or mean) life of a radionuclide is

$$\tau = \frac{1}{N(0)} \int_0^\infty t\,\lambda\,N(t)\,dt = \frac{1}{\lambda} \tag{6}$$

The buildup of a stable decay (daughter) product, which is not initially present, would follow

$$N(0)\left(1-e^{-\lambda t}\right) \tag{7}$$

Activity

The *activity* is the number of decays or disintegrations per unit time [Becquerels (Bq) or Curies (Ci)]

$$A(t) \equiv \lambda\, n(t) = \lambda\, n(0)\, e^{-\lambda t} \tag{8}$$

where a Becquerel is the SI unit defined as one transformation per second, and 1 Ci = 3.7×10^{10} Bq. Figure 1 shows the activity of a (parent) radionuclide where the time scale (ordinate) is expressed in term of the number of half-lives of the radionuclide and the abscissa is measured in comparison to the initial activity (A_0). The *y*-axis could equivalently be stated in terms of $N(t)$ or $n(t)$ as measured in reference to N_0 and n_0, respectively.

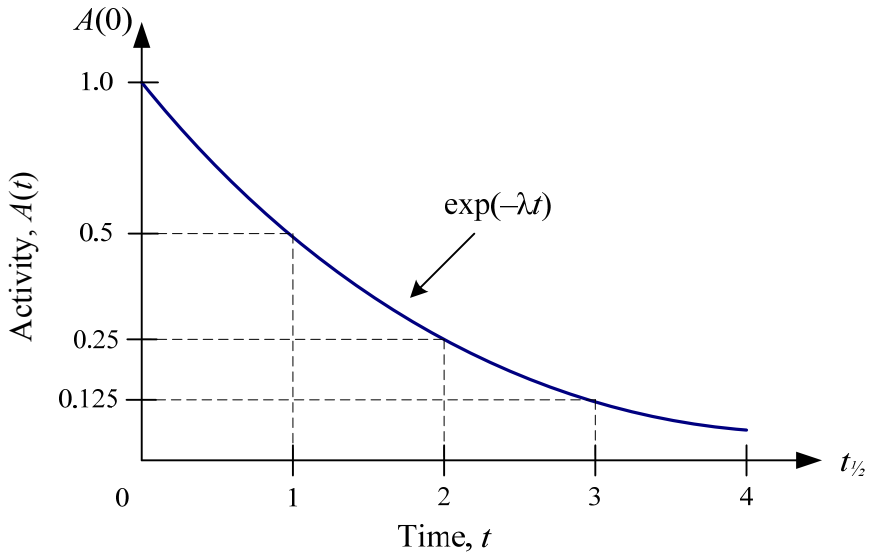


Figure 1. Activity of a radionuclide undergoing simple decay. The radionuclide activity is normalized to the initial activity, $A(0)$, and time is in terms of the number of half-lives.

Example:
Given 1 gram-mole of potassium (K) today, compute: (*a*) the activity today in Bq, and (*b*) the number of K-40 atoms one billion years from now.
Solution:
A gram-mole of any element is 6.022×10^{23} atoms, and in this case equals the number of potassium (K) atoms. Natural potassium is composed of two stable isotopes, K-39 and K-41, and a radioisotope, K-40, which is only 0.0117 atom percent. Hence, today there are

$$n_{\mathrm{K}-40} = (0.000117\ \tfrac{\text{K-40 atoms}}{\text{K atoms}})(6.022\times10^{23}\ \text{K atoms}) = 7.046\times10^{19}\ \text{K - 40 atoms}$$

The half-life of K-40 is 1.277×10^{9} years, such that the decay constant is

$$\lambda = \frac{\ln(2)}{t_{½}} = \frac{\ln(2)}{1.277\times10^{9}\ \text{yr}}\left(\frac{1\,\text{yr}}{365\,\text{day}}\right)\left(\frac{1\,\text{day}}{24\,\text{hr}}\right)\left(\frac{1\,\text{hr}}{3600\,\text{sec}}\right) = 1.721\times10^{-17}\ \tfrac{1}{\text{sec}}$$

(*a*) The activity of the K-40 today is

$$A_0 = \lambda\, n_0 = (1.721\times10^{-17}\ \tfrac{1}{\text{sec}})(7.046\times10^{19}\ \text{atoms}) = 1213\ \text{Bq}$$

(b) The number of K-40 atoms in 10^9 years may be found using Eq. (4):

$$n(t) = n_0\left(\tfrac{1}{2}\right)^{t/t_{½}} = (7.046\times10^{19}\text{ atoms})\left(\tfrac{1}{2}\right)^{10^9\text{ yr}/1.277\times10^9\text{ yr}} = 4.095\times10^{19}\text{ atoms}$$

Specific Activity

The *specific activity* is the activity per unit mass (m) of the radionuclide [Bq/g or Ci/g]

$$SA \equiv \frac{A}{m} = \frac{\lambda n}{m} = \frac{\lambda}{m}\frac{m N_{Av}}{M} = \frac{\lambda N_{Av}}{M} \tag{9}$$

where M is the atomic weight. This expression shows that the specific activity is independent of the actual mass and is a fixed value (*i.e.*, time independent) for a particular radionuclide.

Example:
Compute the specific activity of cobalt-60.
Solution:
The half-life of Co-60 is 5.27 years. The specific activity is computed using Eq. (9):

$$SA_{\text{Co-60}} = \frac{\ln(2)}{(5.27\text{ yrs})}\frac{(6.022\times10^{23}\text{ atoms/g - mole})}{(60\text{ g/g - mole})}\left(\frac{1\text{ yr}}{365\text{ day}}\right)\left(\frac{1\text{ day}}{24\text{ hr}}\right)\left(\frac{1\text{ hr}}{3600\text{ sec}}\right) = 4.19\times10^{13}\ \frac{\text{Bq}}{\text{g}}$$

Effective Half-life

The *effective half-life* takes into account both the radioactive decay and the biological removal of a radioisotope. It is the combination of the radioactive half-life and the biological half-life (like resistors in a parallel circuit).

$$\lambda_{eff} = \lambda_{rad} + \lambda_{bio}$$
$$t_{½,eff} = \frac{\ln(2)}{\lambda_{eff}} = \frac{t_{½,rad}\ t_{½,bio}}{t_{½,rad} + t_{½,bio}} \tag{10}$$

Example:
Iodine-131 has a radioactive half-life of 8 days and a biological half-life of 120 days as it tends to collect in the thyroid. What is the effective half-life of I-131?
Solution:

$$t_{½,eff} = \frac{(8\text{ days})(120\text{ days})}{(8+120\text{ days})} = 7.5\text{ days}$$

Table I. Natural Heavy Decay Chains

Series	Decay Chain	Parent	Parent Half-life (yrs)	Stable End Product
(4*n*+0)	Thorium	$^{232}_{90}$Th	1.405×10^{10}	$^{208}_{82}$Pb
(4*n*+1)	Neptunium	$^{237}_{93}$Np	2.14×10^{6}	$^{209}_{83}$Bi
(4*n*+2)	Uranium	$^{238}_{92}$U	4.468×10^{9}	$^{206}_{82}$Pb
(4*n*+3)	Actinium	$^{235}_{92}$U	7.038×10^{8}	$^{207}_{82}$Pb

Heavy Decay Chains

The natural heavy decay chains consist of four series of radionuclides as summarized in Table I. The ($4n+b$) expression describes the mass number of any member in the series. The numeral "4" occurs because during an alpha transition there is a change in the nucleus of four mass units. The values of "b" (b=0,1,2,3) indicate the number of neutron and/or proton departures from the thorium series ($4n$) where n is an integer. Hence, in the thorium series the parent and each of the daughter products has a mass number perfectly divisible by 4. Note that the series with a parent half-life of $t_{½}>10^{10}$ yrs have decayed very little while those with a half-life of $t_{½}<10^{8}$ yrs are gone. The series still present are detailed in Figure 3. These heavy decay chains undergo compound (serial) and complex (branching) decay schemes, which are explored next.

Compound Decay ($n_1 \rightarrow n_2 \rightarrow n_3$)

The earlier equation for simple decay can be extended to the case in which a radionuclide (n_1) decays to a daughter product (n_2) that is also radioactive, and which subsequently decays to a stable end product (n_3). The differential equation and time domain solution for n_1 are the same as the simple decay situation above.

$$\frac{dn_1}{dt} = -\lambda_1 n_1(t) \quad \Rightarrow \quad s\,n_1(s) - n_1(0) = -\lambda_1 n_1(s) \quad \Rightarrow \quad n_1(t) = n_1(0)\,e^{-\lambda_1 t} \tag{11}$$

The differential equation for n_2, however, includes the production of n_2 from the decay of n_1

$$\frac{dn_2}{dt} = \lambda_1 n_1(t) - \lambda_2 n_2(t) \tag{12}$$

The solution to this differential equation may also be accomplished with Laplace transforms, and substituting for $n_1(s)$ using an expression extracted from Eq. (11) above:

$$\begin{aligned} s\,n_2(s) - n_2(0) &= \lambda_1 n_1(s) - \lambda_2 n_2(s) \\ n_2(s) &= \frac{n_2(0) + \lambda_1 n_1(s)}{(s+\lambda_2)} \\ &= \frac{n_2(0)}{(s+\lambda_2)} + \frac{\lambda_1 n_1(0)}{(s+\lambda_1)(s+\lambda_2)} \end{aligned} \tag{13}$$

Inverse Laplace transforming this expression yields

$$n_2(t) = n_2(0)\,e^{-\lambda_2 t} + \frac{n_1(0)\,\lambda_1}{\lambda_2 - \lambda_1}\left[e^{-\lambda_1 t} - e^{-\lambda_2 t}\right] \tag{14}$$

The differential equation for the end product (granddaughter) n_3 consists only of a production term since there are no losses because the end product is stable.

$$\frac{dn_3}{dt} = \lambda_2 n_2(t) \tag{15}$$

The solution may be found by integrating this expression and substituting Eq. (14)

$$\begin{aligned} n_3(t) &= n_3(0) + \int_0^t \lambda_2\, n_2(\tau)\, d\tau \\ &= n_3(0) + n_2(0)\left(1 - e^{-\lambda_2 t}\right) + n_1(0)\left(1 + \frac{\lambda_2}{\lambda_1 - \lambda_2}\,e^{-\lambda_1 t} - \frac{\lambda_1}{\lambda_1 - \lambda_2}\,e^{-\lambda_2 t}\right) \end{aligned} \tag{16}$$

Alternatively, the solution may be determined using Laplace transforms

$$\frac{dn_3}{dt} = \lambda_2 n_2(t) \quad \Rightarrow \quad s\,n_3(s) - n_3(0) = \lambda_2 n_2(s) \tag{17}$$

Compound Radioactive Decay

There are three cases of interest for compound decay.

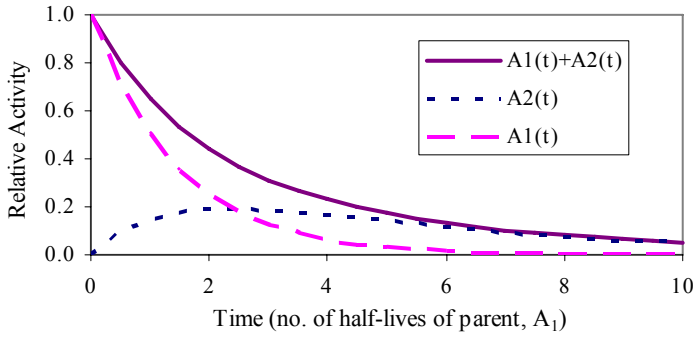
1. The *non-equilibrium* or general case ($\lambda_1>\lambda_2$) requires the use of the full equation (*i.e.*, Eq. (14)) for $n_2(t)$. For $n_2(0)=0$, the full expression is reduced to

$$n_2(t)=\frac{n_1(0)\,\lambda_1}{\lambda_2-\lambda_1}\left[e^{-\lambda_1 t}-e^{-\lambda_2 t}\right] \tag{18}$$

The activities of the parent and daughter are graphed below where it can be seen that eventually the total activity is dominated by the daughter's activity, that is

$$A_1 + A_2 \cong A_2 \qquad \text{for } t \gtrsim 7\,t_{½,1} \tag{19}$$

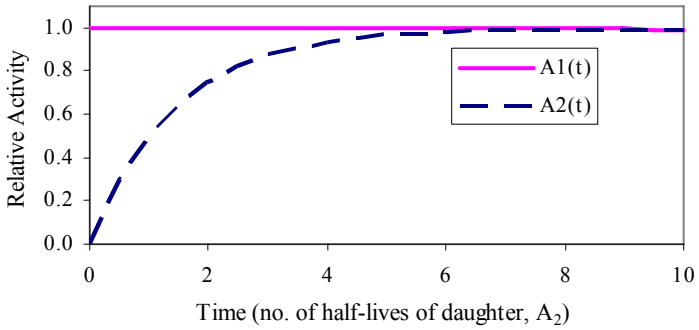
Non-Equilibrium Compound Decay



2. *Secular equilibrium* occurs when the parent is very long-lived compared to the daughter, $\lambda_1<<\lambda_2$. After about seven half-lives of the daughter, the parent's and daughter's activities are equal as shown in the equations and figure below for $n_2(0)=0$

$$n_2(t)\approx\frac{n_1(0)\,\lambda_1}{\lambda_2}\left[1-e^{-\lambda_2 t}\right]$$
$$A_2(t)\equiv\lambda_2\,n_2(t)=\lambda_1\,n_1(0)=A_1(0) \qquad \text{for } t \gtrsim 7\,t_{½,2} \tag{20}$$

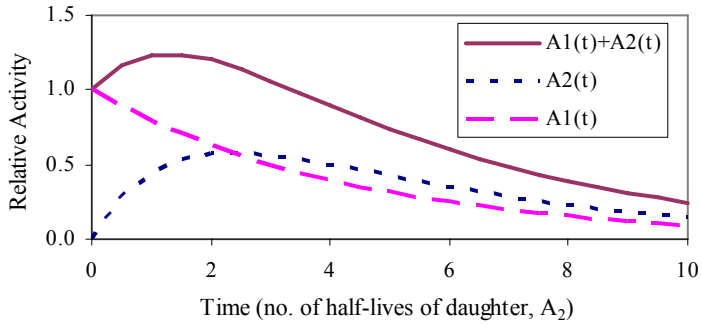
Secular Equilibrium Compound Decay



3. *Transient equilibrium* occurs when the parent is long-lived ($\lambda_1<\lambda_2$) since eventually all the activities decay with the half-life of the parent as illustrated in the equations and figure below for $n_2(0)=0$

$$n_2(t) \approx \frac{n_1(0)\,\lambda_1}{\lambda_2-\lambda_1} e^{-\lambda_1 t} = \frac{\lambda_1}{\lambda_2-\lambda_1} n_1(t)$$
$$A_2(t) \equiv \lambda_2\, n_2(t) = A_1(t) \frac{\lambda_2}{\lambda_2-\lambda_1} \qquad \text{for } t \gtrsim 7\, t_{½,2} \tag{21}$$

Transient Equilibrium Compound Decay



Bateman Equation ($n_1 \rightarrow n_2 \rightarrow n_3 \rightarrow \ldots \rightarrow n_i \rightarrow$)

Bateman developed a general equation for serial decay chains*, such as the heavy decay chains of Th-232, U-235, and U-238. Assuming that the concentrations of all the daughters are initially zero (*i.e.*, $n_i(0)=0$ for $i>1$), the concentration of the i-th radionuclide can be determined from

$$n_i(t) = \lambda_1\,\lambda_2 \cdots \lambda_{i-1}\, n_1(0) \sum_{j=1}^{i} \frac{e^{-\lambda_j t}}{\prod_{\substack{k=1 \\ k\neq j}}^{i} (\lambda_k - \lambda_j)} \tag{22}$$

Example: Natural uranium is composed by atomic percent of 99.2745% U-238, 0.72% U-235, and 0.0055% U-234. Confirm the relative fractions of U-238 and U-234, that is, verify

$$n_{\text{U-238}} / n_{\text{U-234}} = (99.2745)/(0.0055) = 18{,}050$$

Solution: We note that secular equilibrium is eventually established between U-238 and its great-grandchild, U-234, such that their activities are equal:

$$A_{\text{U-238}} = A_{\text{U-234}}$$
$$(\lambda\, n)_{\text{U-238}} = (\lambda\, n)_{\text{U-234}}$$

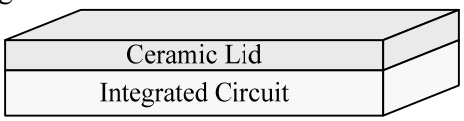
From Figure 3, the half-lives of U-238 and U-234 are 4.468×10^9 yrs and 2.445×10^5 yrs, respectively. Substituting these values into the above expression yields:

$$\frac{n_{\text{U-238}}}{n_{\text{U-234}}} = \frac{\lambda_{\text{U-234}}}{\lambda_{\text{U-238}}} = \frac{\ln(2)/t_{½,\text{U-234}}}{\ln(2)/t_{½,\text{U-238}}} = \frac{t_{½,\text{U-238}}}{t_{½,\text{U-234}}} = \frac{4.468\times10^9 \text{ yrs}}{2.445\times10^5 \text{ yrs}} = 18{,}274$$

Since the fraction of U-234 is only known to two significant figures, the relative fractions are confirmed.

* H. Bateman, "The solution of a system of differential equations occurring in the theory of radio-active transformations," *Proc. Cambridge Phil. Soc.*, 15, p. 423 (1910).

Example: An impurity level of 5 ppm of Th-232 is present in the ceramic packaging material to be used as a direct top covering (lid) for an integrated circuit (IC), as shown below. The ceramic has a density of 4.7 g/cm^3 and an effective atomic weight of 43.5 amu. Determine the maximum alpha flux into the IC.



Solution:
The atomic density of the ceramic is

$$N_{\text{ceramic}} = \frac{\rho\, N_{Av}}{M} = \frac{(4.7\ \text{g/cm}^3)(6.022\times10^{23}\ \text{atoms/g - mole})}{43.5\ \text{g/g - mole}} = 6.51\times10^{22}\ \text{atoms/cm}^3$$

The thorium concentration is

$$N_{\text{Th}} = (5\times10^{-6})\, N_{\text{ceramic}} = (5\times10^{-6})(6.51\times10^{22}) = 3.25\times10^{17}\ \text{atoms/cm}^3$$

The decay constant of Th-232 is

$$\lambda_{\text{Th-232}} = \frac{\ln(2)}{t_{½}} = \frac{\ln(2)}{1.405\times10^{10}\ \text{yr}}\left(\frac{1\ \text{yr}}{365\ \text{d}}\right)\left(\frac{1\ \text{d}}{24\ \text{hr}}\right)\left(\frac{1\ \text{hr}}{3600\ \text{s}}\right) = 1.564\times10^{-18}\ /\,\text{sec}$$

The activity of the thorium is

$$A_{\text{Th-232}} = \lambda\, N = \left(1.564\times10^{-18}\ \tfrac{1}{\text{sec}}\right)\left(3.25\times10^{17}\ \tfrac{\text{atoms}}{\text{cm}^3}\right) = 0.5088\ \text{Bq/cm}^3$$

We must find the depth into the lid from which the alphas will have sufficient energy to escape the lid, that is, we determine an active region of the lid in terms of an alpha range into the lid. We find that Th-232 emits alphas at two different energies: 4.016 MeV (77%) and 3.957 MeV (23%). The range of these ~ 4 MeV alphas in air is

$$R_{air} = 1.24\, E_\alpha - 2.62 = (1.24)(4\ \text{MeV}) - 2.62 = 2.34\ \text{cm}$$

The corresponding range in the ceramic lid is computed using the Bragg-Kleeman rule

$$R = 2.3\times10^{-4}\,\frac{\sqrt{M}}{\rho}\,R_{air} = \left(2.3\times10^{-4}\right)\left(\frac{\sqrt{43.5}}{4.7}\right)(2.34\ \text{cm}) = 0.000755\ \text{cm} = 7.55\ \mu\text{m}$$

Assuming a one-dimensional geometry, only half the alphas at the most move toward the IC such that the maximum alpha emission flux into the IC is

$$\phi_\alpha = \tfrac{1}{2}\,A\,d = \frac{1}{2}\left(0.6115\,\frac{\text{Bq}}{\text{cm}^3}\right)(0.000755\ \text{cm})\left(\frac{1\ \text{alpha/sec}}{\text{Bq}}\right) = 0.00023\,\frac{\text{alphas}}{\text{cm}^2\cdot\text{sec}}$$

Of course, a thinner lid than 7.55 μm would emit fewer alphas. The above analysis does not include any of the alpha emissions from progeny of Th-232.

Complex Radioactive Decay

A radionuclide may also decay by multiple means, for example, by both α and β decay. As examples, Figure 3 shows two such complex or branching decay schemes: (1) in the Th-232 series, Bi-212 decays to either Po-212 or Tl-208, and (2) in the U-235 series, Ac-227 decays to Th-227 or Fr-223

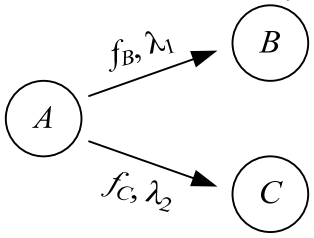


Figure 2. Complex decay scheme in which radionuclide *A* decays to either *B* or *C*.

Suppose that radionuclide A decays proportionally to B and C according to fractions f_B and f_C, respectively, as depicted in Figure 2. Since λ_A represents the decay probability for A, then the probability of decay from A to B is $\lambda_1 = f_B\ \lambda_A$, and likewise to C is $\lambda_2 = f_C\ \lambda_A$. The overall decay probability λ_A is the sum of the individual probabilities, that is, $\lambda_A = \lambda_1 + \lambda_2$ (*i.e.*, a joint probability from the union of the two decay paths). The balance equation for radionuclide A is therefore

$$\frac{dn_A}{dt} = -\lambda_A\, n_A(t) = -(\lambda_1 + \lambda_2)\, n_A(t) \tag{23}$$

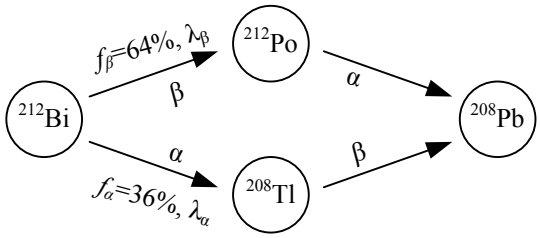
The activity of A can be shown to be

$$A_A(t) = \lambda_A\, n_A(t) = (\lambda_1 + \lambda_2)\, n_A(t) = A_1(t) + A_2(t)$$
$$\text{and} \tag{24}$$
$$A_A(t) = A_A(0)\, e^{-\lambda_A t} = A_A(0)\, e^{-(\lambda_1+\lambda_2)t} = \lambda_A\, n_A(0)\, e^{-(\lambda_1+\lambda_2)t}$$

If B and C are stable, then intuitively from Eq. (7):

$$\begin{aligned} n_B(t) &= n_B(0) + f_B\, n_A(0)\left(1 - e^{-\lambda_A t}\right) \\ n_C(t) &= n_C(0) + f_C\, n_A(0)\left(1 - e^{-\lambda_A t}\right) \end{aligned} \tag{25}$$

Example: The decay of Bi-212 involves a complex decay scheme whose daughters both decay to the same grandchild (stable Pb-208) as illustrated below. Determine whether the time to decay from Bi-212 to Pb-208 differs based on the decay branch taken.



Solution: We begin by finding the decay constant of Bi-212

$$\lambda_{\text{Bi-212}} = \ln(2) / t_{½} = \ln(2) / (60.55\ \text{min}) = 0.01145\ /\text{min}$$

Next, determine the decay constants associated with the initial two (α and β) decay branches

$$\lambda_\beta = f_\beta\ \lambda_{\text{Bi-212}} = (0.6407)(0.01145\ /\text{min}) = 0.00734\ /\text{min}$$

$$\lambda_\alpha = f_\alpha\ \lambda_{\text{Bi-212}} = (0.3593)(0.01145\ /\text{min}) = 0.00411\ /\text{min}$$

From Figure 3, the half-lives of Po-212 and Tl-208 are 305 ns and 3.07 min, respectively. Using Eq. (6), the corresponding average lives ($\tau = 1/\lambda = t_{½} / \ln(2)$) of the radionuclides are

Upper branch:
$$\tau_{\text{Bi-212},\beta} = 1/\lambda_\beta = 1/(0.00734\ /\text{min}) = 136.2\ \text{min}$$
$$\tau_{\text{Po-212}} = t_{1/2} / \ln(2) = (305\ \text{ns}) / \ln(2) = 440\ \text{ns}$$

Lower branch:
$$\tau_{\text{Bi-212},\alpha} = 1/\lambda_\alpha = 1/(0.00411\ /\text{min}) = 243.3\ \text{min}$$
$$\tau_{\text{Tl-208}} = t_{1/2} / \ln(2) = (3.07\ \text{min}) / \ln(2) = 4.43\ \text{min}$$

This implies that the average time in the upper (Bi-212→Po-212→Pb-208) branch is shorter than in the Bi-212→Tl-208→Pb-208 path. Noteworthy is that both of these paths represent secular equilibrium behavior.

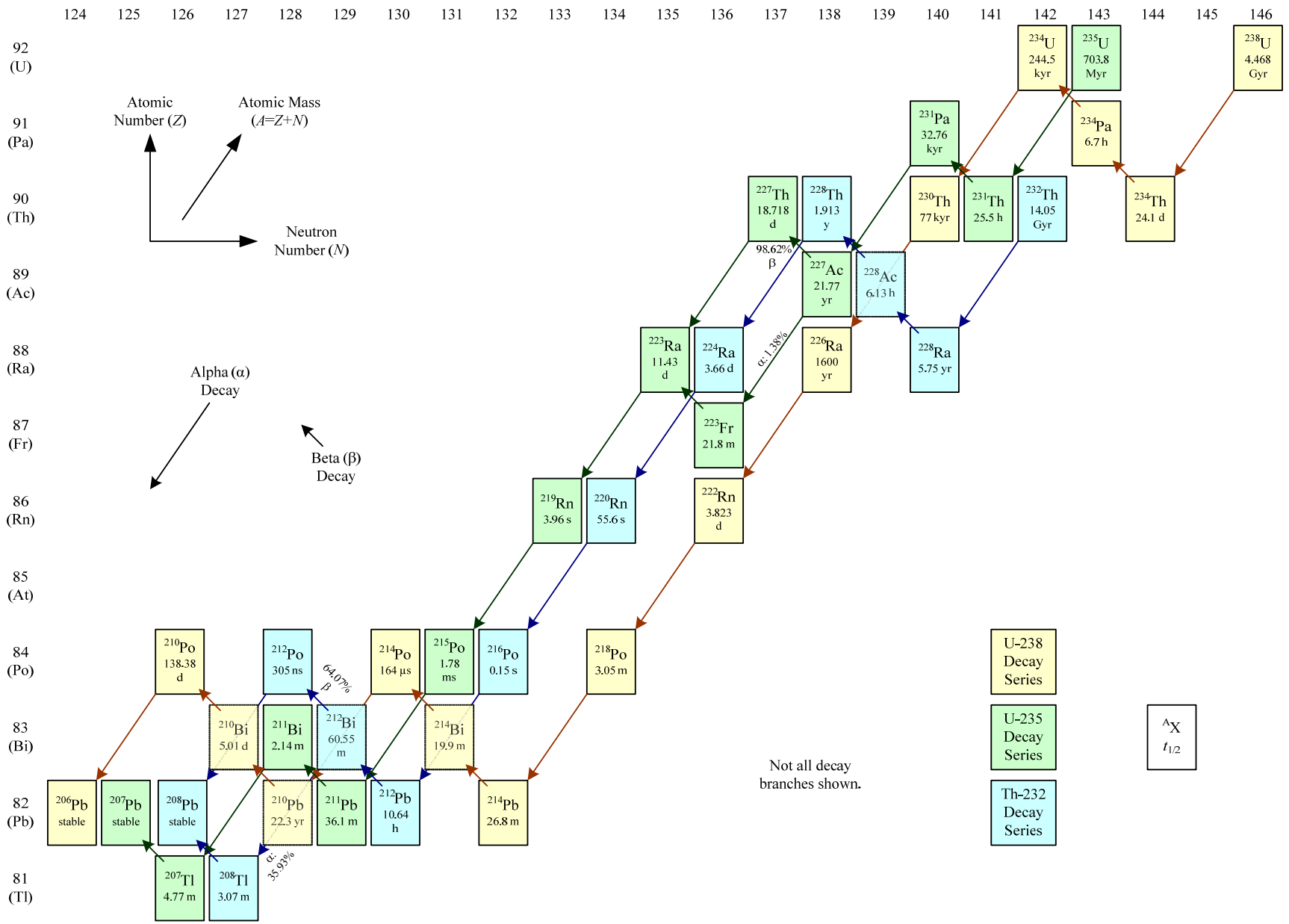


Figure 3. Thorium-232, Uranium-235 and U-238 decay chains referenced to atomic (left) and neutron (top) numbers.