

Equations of Radioactive Decay of Isotopes in a Decay Chain

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Abstract

In this document I show how to solve the coupled differential equations which describe the decay of isotopes in a decay chain. In this paper the initial condition is chosen as only the parent isotope existing at $t = 0$. If a daughter isotope in a decay chain has a significantly shorter decay time constant than the parent it is possible to arrive at secular equilibrium where the decay rates of the daughter and parent isotopes are equal. For the more realistic case of approximate secular equilibrium I calculate the ratio of the parent to daughter decay rates (equilibrium constants). I show how this works specifically in the decay chain of $^{227}_{89}\text{Ac}$ down to the stable isotope $^{207}_{82}\text{Pb}$. At equilibrium one would expect a total of 5 alphas for each Ac-227 decay, but one actually gets 5.015 alphas per Ac-227 decay.

Keywords: half-life, decay chain, isotopes

1 Coupled Equations of a Decay Chain

In scientific research radioactive isotopes are often utilized both as a source for calibrating instruments and as a subject of study themselves. It is desirable to know the equations governing the decay of a given set of isotopes. For a single isotope that has no parent isotopes in the material of interest, the number of atoms of that isotope is given by the simple and familiar decay equation:

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

which has the simple solution

$$N(t) = N(0)e^{-\lambda t} \quad (2)$$

where λ is the decay constant and $N(0)$ represents the number of atoms of the isotope at $t = 0$.

In a decay chain the number of atoms of each isotope depends on the rate of production of the parents as well as the rate of decay so that the rate of decay of each isotope depends upon the decay rate of all isotopes above it in the chain. This can be described by the following set of coupled differential equations:

$$\begin{aligned} \dot{N}_0(t) &= -\lambda_0 N_0(t) \\ \dot{N}_1(t) &= -\lambda_1 N_1(t) + \lambda_0 N_0(t) \\ &\vdots \\ \dot{N}_n(t) &= -\lambda_n N_n(t) + \lambda_{n-1} N_{n-1}(t) \end{aligned} \quad (3)$$

These equations can be solved in order using an integrating factor. An ODE of the form

$$\frac{dy(t)}{dt} + p(t)y(t) = q(t), \quad (4)$$

can be solved using the integrating factor $e^{\int^t p(t')dt'}$ giving the following solution:

$$y(t) = \frac{\int e^{\int^t p(t')dt'} q(t) dt + c}{e^{\int^t p(t')dt'}}. \quad (5)$$

If we assume that at $t = 0$ we start with only the parent isotope, Eq. 5 together with Eq. 2 yields

$$N_1(t) = \lambda_0 N_0(0) \left(\frac{e^{-\lambda_0 t} - e^{-\lambda_1 t}}{\lambda_1 - \lambda_0} \right).$$

Inserting this into the equation for $N_2(t)$ and solving similarly gives

$$N_2(t) = \lambda_0 \lambda_1 N_0(0) \left(\frac{e^{-\lambda_0 t} - e^{-\lambda_2 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_0)} - \frac{e^{-\lambda_1 t} - e^{-\lambda_2 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_1)} \right).$$

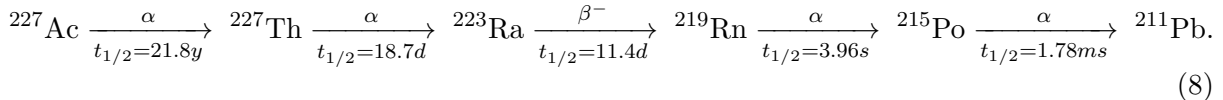
For $N_3(t)$ this gives

$$N_3(t) = \lambda_0 \lambda_1 \lambda_2 N_0(0) \left[\frac{e^{-\lambda_0 t} - e^{-\lambda_3 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_0)(\lambda_3 - \lambda_0)} - \frac{e^{-\lambda_1 t} - e^{-\lambda_3 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} \right. \\ \left. - \frac{e^{-\lambda_2 t} - e^{-\lambda_3 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_0)(\lambda_3 - \lambda_2)} + \frac{e^{-\lambda_2 t} - e^{-\lambda_3 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} \right]. \quad (6)$$

For $N_4(t)$ this gives

$$N_4(t) = \alpha \left[\frac{e^{-\lambda_0 t} - e^{-\lambda_4 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_0)(\lambda_3 - \lambda_0)(\lambda_4 - \lambda_0)} - \frac{e^{-\lambda_1 t} - e^{-\lambda_4 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)(\lambda_4 - \lambda_1)} \right. \\ - \frac{e^{-\lambda_2 t} - e^{-\lambda_4 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_0)(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)} + \frac{e^{-\lambda_2 t} - e^{-\lambda_4 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)} \\ - \frac{e^{-\lambda_3 t} - e^{-\lambda_4 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_0)(\lambda_3 - \lambda_0)(\lambda_4 - \lambda_3)} - \frac{e^{-\lambda_3 t} - e^{-\lambda_4 t}}{(\lambda_0 - \lambda_1)(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)(\lambda_4 - \lambda_3)} \\ \left. + \frac{e^{-\lambda_3 t} - e^{-\lambda_4 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_0)(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_3)} - \frac{e^{-\lambda_3 t} - e^{-\lambda_4 t}}{(\lambda_1 - \lambda_0)(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_3)} \right], \quad (7)$$

where $\alpha = \lambda_0 \lambda_1 \lambda_2 \lambda_3 N_0(0)$. Each successive equation in the decay chain becomes more complicated with 2^{n-1} terms; however, often only one or a few terms dominate the expression as time progresses. Take for example the decay chain of ^{227}Ac to ^{211}Pb as follows:



Using $\lambda_0 = \lambda_{Ac} = 1.01 \times 10^{-9} \text{ s}^{-1}$, $\lambda_1 = \lambda_{Th} = 4.29 \times 10^{-7} \text{ s}^{-1}$, $\lambda_2 = \lambda_{Ra} = 7.02 \times 10^{-7} \text{ s}^{-1}$, and $\lambda_3 = \lambda_{Rn} = 0.175 \text{ s}^{-1}$, we can approximate $N_3(t)$ as follows:

$$N_3(t) \approx \lambda_0 \lambda_1 \lambda_2 N_0(0) \left[\frac{e^{-\lambda_0 t} - e^{-\lambda_3 t}}{\lambda_1 \lambda_2 \lambda_3} - \frac{e^{-\lambda_1 t} - e^{-\lambda_3 t}}{\lambda_1 (\lambda_2 - \lambda_1) \lambda_3} - \frac{e^{-\lambda_2 t} - e^{-\lambda_3 t}}{\lambda_1 \lambda_2 \lambda_3} - \frac{e^{-\lambda_2 t} - e^{-\lambda_3 t}}{\lambda_1 (\lambda_2 - \lambda_1) \lambda_3} \right] \quad (9)$$

$$= \frac{\lambda_0}{\lambda_3} N_0(0) \left[\left(e^{-\lambda_0 t} - e^{-\lambda_2 t} \right) - \frac{\lambda_2 (e^{-\lambda_1 t} + e^{-\lambda_2 t} - 2e^{-\lambda_3 t})}{(\lambda_2 - \lambda_1)} \right] \quad (10)$$

With $\lambda_3 = 0.175 \text{ s}^{-1}$, the last term involving $e^{-\lambda_3 t}$ becomes negligible in less than a minute leaving

$$\begin{aligned} N_3(t) &\approx \frac{\lambda_0}{\lambda_3} N_0(0) \left[\left(e^{-\lambda_0 t} - e^{-\lambda_2 t} \right) - \frac{\lambda_2 (e^{-\lambda_1 t} + e^{-\lambda_2 t})}{(\lambda_2 - \lambda_1)} \right] \\ &= \frac{t_{1/2}^{(3)}}{t_{1/2}^{(0)}} N_0(0) \left[\left(e^{-\lambda_0 t} - e^{-\lambda_2 t} \right) - \frac{\lambda_2 (e^{-\lambda_1 t} + e^{-\lambda_2 t})}{(\lambda_2 - \lambda_1)} \right] \end{aligned} \quad (11)$$

Since the terms involving $e^{-\lambda_1 t}$ and $e^{-\lambda_2 t}$ will decay much faster than the $e^{-\lambda_0 t}$ term, after 10 half-lives of ^{227}Th (about a half a year), the equation will be dominated simply by the first term which is the decay of ^{227}Ac , that is, for $t > \sim 180 \text{ d}$

$$N_3(t) \approx \frac{\lambda_0}{\lambda_3} N_0(0) e^{-\lambda_0 t} = \frac{t_{1/2}^{(3)}}{t_{1/2}^{(0)}} N_0(0) e^{-\lambda_0 t} \quad (12)$$

Figure 1 compares Equations 11 (red) and 12 (blue) to demonstrate how quickly this approximation of Eq. 12 becomes valid. After ~ 180 days the decay is completely dominated by the Actinium decay rate.

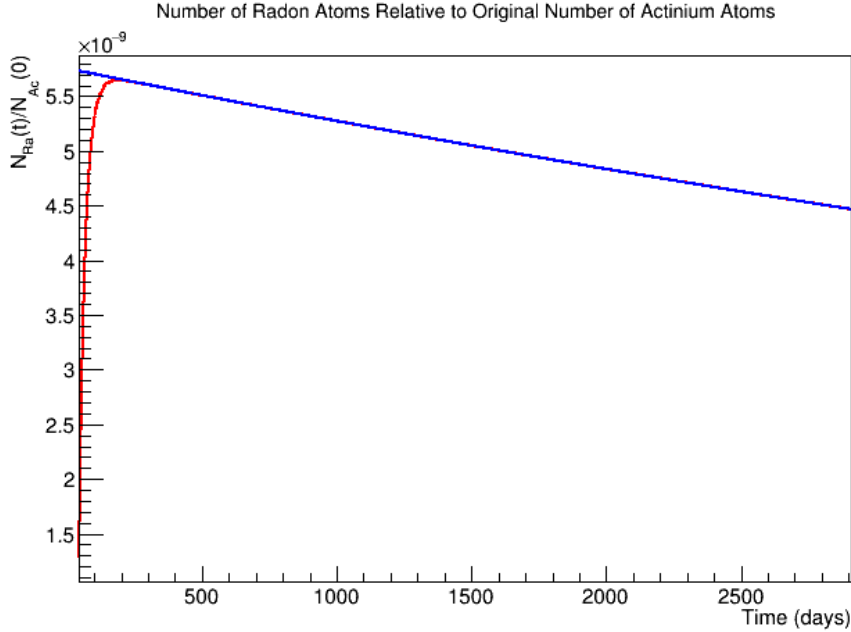


Figure 1: Comparison of decay equations given in Equations 11 (red) and 12 (blue) showing how quickly the approximate solution (blue) becomes valid.

2 Secular Equilibrium

Secular equilibrium occurs between two given isotopes when the decay rates to and from that isotope are equal. This occurs for the decay chain in Eq. 3 when the decay rate of ^{227}Ac equals

that of ^{219}Rn which implies that the ratio of concentrations of the two isotopes is approximately constant. We can see from Eq. 11 that this state is reached when the ratio of the relative concentrations of the isotopes is equal to the ratio of the half-lives of the isotopes. That is, at equilibrium the following relationship holds:

$$\frac{N_{Rn}(t)}{N_{Ac}(t)} \approx \frac{\lambda_{Ac}}{\lambda_{Rn}} = \frac{t_{1/2}^{(Rn)}}{t_{1/2}^{(Ac)}} \approx 5.76 \times 10^{-9}.$$

Perhaps a simpler way of looking at this is to realize that at equilibrium the decay rate of

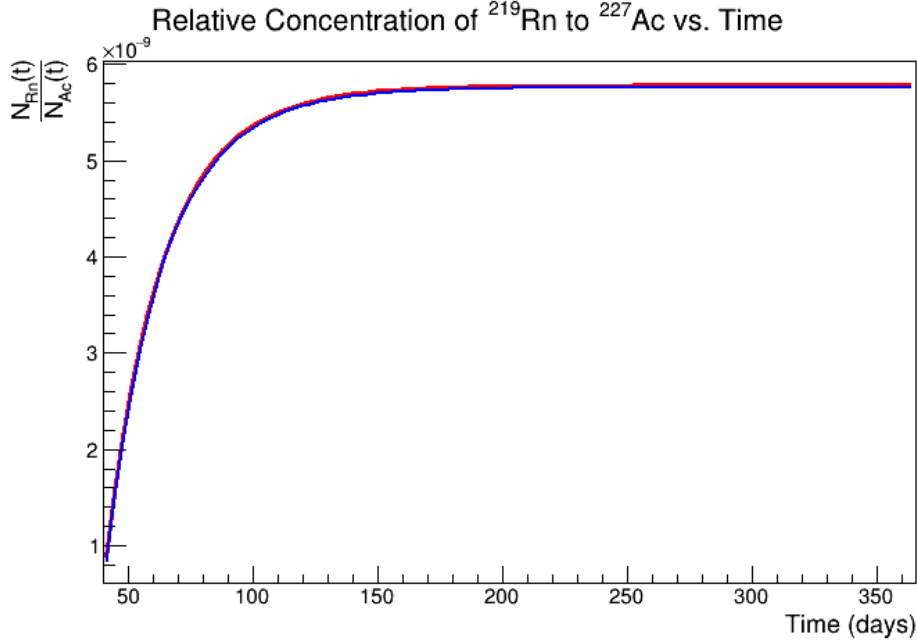


Figure 2: Relative concentration of ^{219}Ra to ^{227}Ac versus time demonstrating that equilibrium is reached after ~ 180 days. The red curve is the exact solution given by the ratio of Eq. 6 to Eq. 2. The approximate solution (blue) is given by the ratio of Eq. 11 to Eq. 2.

Ac-227 and Rn-219 must be equal. The decay rate of an isotope as a function of time is given by $\frac{dN(t)}{dt} = -\lambda N_0 e^{-\lambda t}$, where λ is the lifetime of the isotope and N_0 is the number of atoms at $t = 0$. Since the decay rates are equal we have

$$\lambda_{Ac} N_0^{(Ac)} e^{-\lambda_{Ac} t} = \lambda_{Rn} N_0^{(Rn)} e^{-\lambda_{Rn} t} \longrightarrow \frac{N_0^{(Ac)} e^{-\lambda_{Ac} t}}{t_{1/2}^{(Ac)}} = \frac{N_0^{(Rn)} e^{-\lambda_{Rn} t}}{t_{1/2}^{(Rn)}} \quad (13)$$

For $t \ll t$ i.e. the system is in secular equilibrium at $t = 0$, we have

$$\frac{N_0^{(Ac)}}{t_{1/2}^{(Ac)}} = \frac{N_0^{(Rn)}}{t_{1/2}^{(Rn)}}.$$

3 Approximate Secular Equilibrium and Equilibrium Constants

¹ In the case where the half-life of the parent isotope is much longer than the daughters, secular equilibrium can be approximately reached. However, secular equilibrium is only approximate when considering a set amount of the parent isotope since the activity of each isotope eventually decays away at the half-life of the parent. For decay chains such as Ac-227 with the parent isotope having a longer half-life (by factors of 100-1000) than the daughters a small correction must be applied to the equilibrium value to correctly calculate the activity of the daughters. Intuitively, this can be thought of as coming from the fact that it takes the system (decay chain) a finite amount of time to adjust to the reduced parent activity. Thus, after approximate equilibrium is reached, the activity of the daughters at any given time is greater than that of the parent isotope. The ratio of the daughter activity to that of the parent is called the “equilibrium constant”.

To see this first we can define the activity A_i of a given isotope i using Eq 3 as $A_i(t) = \lambda_i N_i(t)$. For the parent isotope ($i = 0$) this becomes

$$A_0(t) = \lambda_0 N_0(0) e^{-\lambda_0 t} \equiv A_0(0) e^{-\lambda_0 t}. \quad (14)$$

The activity of the first daughter is likewise given by

$$A_1(t) = \lambda_0 \lambda_1 N_0(0) \left(e^{-\lambda_0 t} - e^{-\lambda_1 t} \right) = A_0(0) \frac{\lambda_1}{\lambda_1 - \lambda_0} \left(e^{-\lambda_0 t} - e^{-\lambda_1 t} \right). \quad (15)$$

The equilibrium constant or ratio of the activity of the daughter to the parent is given by

$$k_1 = \frac{A_1(t)}{A_0(t)} = \frac{\lambda_1}{\lambda_1 - \lambda_0} \left(\frac{e^{-\lambda_0 t} - e^{-\lambda_1 t}}{e^{-\lambda_0 t}} \right) = \frac{\lambda_1}{\lambda_1 - \lambda_0} \left(1 - e^{-(\lambda_1 - \lambda_0)t} \right). \quad (16)$$

After several of the (comparatively short) daughter half-lives this goes to

$$k_1 \xrightarrow{t \gg t_{1/2}^{(1)}} \frac{\lambda_1}{\lambda_1 - \lambda_0} = \frac{t_{1/2}^{(0)}}{t_{1/2}^{(0)} - t_{1/2}^{(1)}} \quad (17)$$

Likewise for the activity of the second generation we find

$$A_2(t) = \lambda_0 N_0(0) \frac{\lambda_1 \lambda_2}{\lambda_1 - \lambda_0} \left(\frac{e^{-\lambda_0 t} - e^{-\lambda_1 t}}{\lambda_2 - \lambda_0} - \frac{e^{-\lambda_1 t} - e^{-\lambda_2 t}}{\lambda_2 - \lambda_1} \right) = A_0(0) \frac{\lambda_1 \lambda_2}{\lambda_1 - \lambda_0} \left(e^{-\lambda_0 t} - e^{-\lambda_1 t} \right). \quad (18)$$

Once again we take the ratio of the activity of the second generation to the parent to find the equilibrium constant.

$$k_2 = \frac{A_2(t)}{A_0(t)} = \frac{\lambda_1 \lambda_2}{\lambda_1 - \lambda_0} \left(\frac{e^{-\lambda_0 t} - e^{-\lambda_1 t}}{\lambda_2 - \lambda_0} - \frac{e^{-\lambda_1 t} - e^{-\lambda_2 t}}{\lambda_2 - \lambda_1} \right) / e^{-\lambda_0 t}. \quad (19)$$

After several daughter half-lives the second term in the parentheses becomes negligible and the first term goes to $1/(\lambda_2 - \lambda_0)$ giving

$$k_2 \xrightarrow{t \gg t_{1/2}^{(2)}} \frac{\lambda_1}{\lambda_1 - \lambda_0} \frac{\lambda_2}{\lambda_2 - \lambda_0} = \frac{t_{1/2}^{(0)}}{t_{1/2}^{(0)} - t_{1/2}^{(1)}} = k_1 \frac{t_{1/2}^{(0)}}{t_{1/2}^{(0)} - t_{1/2}^{(2)}}. \quad (20)$$

¹This derivation follows that of <http://www.med.harvard.edu/jpnm/physics/nmltd/radprn/sect2/2.2/2.2.3.html>.

In general we can show that k_i is given recursively by

$$k_i = k_{i-1} \frac{t_{1/2}^{(0)}}{t_{1/2}^{(0)} - t_{1/2}^{(i)}}, \quad x = 1, 2, \dots$$

The net effect of this non-equilibrium correction on the activity of the daughter isotopes in the Ac-227 decay chain is to give a slightly higher decay rate for daughters than for the parent, Ac-227. The non-equilibrium constants for the main decay branches of Ac-227 are calculated in [1]. Table 1 in this paper gives a nice summary of the results. I was able to reproduce this table using the probabilities listed and the half-life values from the online interactive IAEA chart of the nuclides. Summing all the alpha decays from this table for each decay of the parent Ac-227 we would expect about 5.015 total alpha decays (parent and daughters included).

References

- [1] K. Kossert, K. Bokeloh, R. Dersch, and O. Nähle. Activity determination of 227ac and 223ra by means of liquid scintillation counting and determination of nuclear decay data. *Applied Radiation and Isotopes*, 95:143 – 152, 2015.