

THE ATOMIC NUCLEUS

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CHAPTER 15

Radioactive-series Decay

The exponential laws which govern the decay and growth of radioactive substances were first formulated by Rutherford and Soddy in 1902 (R52) in order to explain their experiments on the thorium series of radioactive substances. Useful mathematical generalizations were made in 1910 by Bateman (B20). The more general forms of the decay and growth equations are therefore often referred to as "the Bateman equations." For three decades the applicability of the equations governing series decay was confined to the uranium, actinium, and thorium series of naturally occurring radioactive substances. With the discovery of nuclear fission a vast number of cases of radioactive-series decay appeared among the fission products. The behavior of all these can be understood with the help of the original Bateman equations.

1. Decay of a Single Radioactive Nuclide

a. Radioactive Decay Constant λ . Consider a group containing a large number A of identical radioactive atoms. Let the probability that any particular atom will disintegrate in unit time be λ , the total *radioactive decay constant*. Then the *activity* of these atoms, i.e., the total number of disintegrations per unit time (in a time which is short compared with $1/\lambda$), will be simply $A\lambda$. The rate of depletion, dA/dt , of the group of atoms is equal to the activity, so long as we do not provide any new supply of radioactive atoms. Because A decreases as time increases, we insert a minus sign and write

$$\frac{dA}{dt} = -A\lambda \quad (1.1)$$

Rewriting this fundamental relationship in integral form, with the variables separated, we have

$$\int \frac{dA}{A} = - \int \lambda dt \quad (1.2)$$

We now make the fundamental assumption that *the probability of decay of an atom is independent of the age of that atom*. Then if λ is independent of t and is constant, we can integrate Eq. (1.2) simply. If

there were A_0 atoms at $t = 0$, while A atoms remain at time $t = t$, then Eq. (1.2) becomes

$$\ln \left(\frac{A}{A_0} \right) = -\lambda t \quad (1.3)$$

or, in the usual exponential form,

$$A = A_0 e^{-\lambda t} \quad (1.4)$$

Recalling that $A_0 \lambda$ is the activity at $t = 0$, Eq. (1.4) can be written in terms of the ratio of the activities at t and at $t = 0$. Because λ is independent of t , we can write

$$\frac{A}{A_0} = e^{-\lambda t} \quad (1.5)$$

in agreement with the empirical law of radioactive decay and the associated disintegration hypothesis of Rutherford and Soddy (R52).

The exponential law, Eq. (1.4), of radioactive decay can also be derived from the laws of chance† without any knowledge of the mechanism of disintegration. The only assumptions needed are that:

1. The probability of decay is the same for all atoms of the species.
2. The probability of decay is independent of the age of the particular atom.

These conditions are mathematically "sufficient," as was first shown by von Schweidler (S20). They are also mathematically "necessary" conditions, as is most readily seen from the experimental fact that the statistical fluctuations in α -ray emission obey the Poisson distribution (Chap. 26), for the derivation of which assumptions analogous to those given above are both necessary and sufficient. Ruark (R38) has developed an analytical proof that these conditions are mathematically necessary. The point is of importance because the wave-mechanical theories of α -ray and β -ray radioactive decay involve the basic assumption that the probability λ of decay in unit time is independent of the age of the particular atom in question.

The most direct experimental proof that the decay constants of a number of naturally occurring radioactive substances have not changed in the last 10^9 yr comes from the sharpness of individual rings in uranium and thorium pleochroic halos in mica (H32, H33, H34).

The disintegration law of Eq. (1.4) applies universally to all radio-

† If ϑ is a time interval which is very small compared with $1/\lambda$, then $\lambda\vartheta$ is the chance that a particular atom will decay in the time ϑ . Its chance of survival for a time ϑ is then $(1 - \lambda\vartheta)$, for a time 2ϑ is $(1 - \lambda\vartheta)^2$, . . . , and for any arbitrary time $t = n\vartheta$ is $(1 - \lambda\vartheta)^n = (1 - \lambda\vartheta)^{t/\vartheta}$. It can be verified easily by series expansion that, if the arbitrary time interval ϑ is taken as very small, then in the limit of $\vartheta/t \rightarrow 0$

$$\lim_{\vartheta/t \rightarrow 0} (1 - \lambda\vartheta)^{t/\vartheta} = e^{-\lambda t}$$

Thus the chance of survival is $e^{-\lambda t}$ for each atom. Hence the average fraction which survives a time t is also $e^{-\lambda t}$. We must expect statistical fluctuations in the actual fraction which survives, such that the average value is $e^{-\lambda t}$.

active nuclides, but the constant λ is different for each nuclide. The known radioactive nuclides extend between $\lambda = 3 \times 10^6 \text{ sec}^{-1}$ (for ThC') and $\lambda = 1.58 \times 10^{-18} \text{ sec}^{-1}$ (for Th), a range of over 10^{24} . The decay constant λ is one of the most important characteristics of each radioactive nuclide; it is essentially independent of all physical and chemical conditions such as temperature, pressure, concentration, or age of the radioactive atoms. Among the more than 800 known radioactive nuclides, no two have exactly the same decay constant. The identification of some radioactive samples can be made simply by measuring λ , which can serve as a type of qualitative chemical analysis.

b. Partial Decay Constants. Many nuclides have at risk several alternative modes of decay. For example, Cu^{64} can decay by electron capture or by positron β -ray emission or by negatron β -ray emission. If the competing modes of decay of any nuclide have probabilities $\lambda_1, \lambda_2, \lambda_3, \dots$ per unit time, then the total probability of decay is represented by the total decay constant λ , where

$$\lambda = \lambda_1 + \lambda_2 + \lambda_3 + \dots \quad (1.6)$$

The "partial activity" of a sample of A nuclei, if measured by a particular mode of decay characterized by λ_i , is then

$$\frac{dA_i}{dt} = \lambda_i A = \lambda_i A_0 e^{-\lambda t} \quad (1.7)$$

and the total activity is

$$\frac{dA}{dt} = \sum_i \frac{dA_i}{dt} = A \sum_i \lambda_i = \lambda A_0 e^{-\lambda t} \quad (1.8)$$

Note that partial activities, such as positron β rays from Cu^{64} , are proportional to total activities at all times. Each partial activity falls off with time as $e^{-\lambda t}$, not as $e^{-\lambda_i t}$. Physically, this is because the decrease of activity with time is due to the depletion of the stock of atoms A , and this depletion is accomplished by the combined action of all the competing modes of decay.

c. Units of Radioactivity. The curie unit was redefined in 1950 by action of the international Joint Commission on Standards, Units, and Constants of Radioactivity (P3) in such a way that it applies to all radioactive nuclides and is no longer tied to the presumed activity of 1 g of radium. This definition is "*The curie is a unit of radioactivity defined as the quantity of any radioactive nuclide in which the number of disintegrations per second is 3.700×10^{10} .*" The "number of disintegrations" is the sum of all competing modes of disintegration. Therefore the full decay scheme of a nuclide has to be known, including the electron-capture branching, before the quantity of any sample can be expressed in curies as a result of measurements on any particular mode of disintegration, such as β rays or α rays. In practical use, the "quantity of" any radionuclide is usually nearly synonymous with the "total activity of" the nuclide.

d. Half-period T . The half-period T is the time interval over which the chance of survival of a particular radioactive atom is exactly one-half. Then, if λ is the total decay constant, Eq. (1.3) gives

$$T = \frac{\ln 2}{\lambda} = \frac{0.693}{\lambda} \quad (1.9)$$

In a large initial stock of A_0 atoms, with initial activity $A_0\lambda$, the expectation value of the activity A , one half-period later, is $A = A_0\lambda/2$.

For mnemonic reasons, the half-period T (or $T_{\frac{1}{2}}$ whenever there is any ambiguity about symbols) is much more frequently employed than the decay constant λ . The *half-period* is sometimes also called the *half-value time* or, with less justification, the *half-life*.

Two related periods which are useful in the laboratory are the nine-tenths period and the one-tenth period. Nine-tenths of the atoms survive longer than the *nine-tenths period* $T_{\frac{9}{10}}$ whose value is

$$T_{\frac{9}{10}} = \frac{\ln(\frac{10}{9})}{\lambda} = 0.1520T_{\frac{1}{2}} \simeq \frac{3}{20} T_{\frac{1}{2}} \quad (1.10)$$

One-tenth of the atoms survive longer than the *one-tenth period*, given by

$$T_{\frac{1}{10}} = \frac{\ln 10}{\lambda} = 3.322T_{\frac{1}{2}} \simeq \frac{10}{3} T_{\frac{1}{2}} \quad (1.11)$$

Cruder but handier approximations are $T_{\frac{9}{10}} < \frac{1}{8}T_{\frac{1}{2}}$ and $T_{\frac{1}{10}} > 3T_{\frac{1}{2}}$.

e. Mean Life τ . The actual life of any particular atom can have any value between 0 and ∞ . The average life of a large number of atoms is, however, a definite and important quantity.

If there are A_0 atoms present initially, then the number remaining undecayed at a subsequent time t is $A = A_0e^{-\lambda t}$. Each of these atoms has a life longer than t . Those which decay between t and $t + dt$ each have a life span t . The absolute number of atoms having a life t is therefore $A dt = A_0\lambda e^{-\lambda t} dt$. The total lifetime L of all the atoms is therefore

$$L = \int_0^{\infty} tA\lambda dt = \int_0^{\infty} tA_0\lambda e^{-\lambda t} dt = \frac{A_0}{\lambda} \quad (1.12)$$

Then the average lifetime L/A_0 , which is called the *mean life* τ , is simply

$$\tau = \frac{1}{\lambda} \quad (1.13)$$

Thus the mean life exceeds the half-period and is always given by

$$\tau = \frac{T}{0.693} = 1.44T \quad (1.14)$$

Substitution in Eq. (1.4) shows that the mean life is the time required for the number of atoms, or their activity, to fall to $e^{-1} = 0.368$ of any initial value.

f. Total Number of Radioactive Atoms. The total number of radioactive atoms A present at any time is simply the product of the total activity $A\lambda$ and the mean life, because

$$(A\lambda)\tau = \frac{A\lambda}{\lambda} = A \quad (1.15)$$

The relationships of A_0 , A , t , λ , T , and τ are illustrated graphically in Fig. 1.1. It will be noted that the area under the decay curve of activity

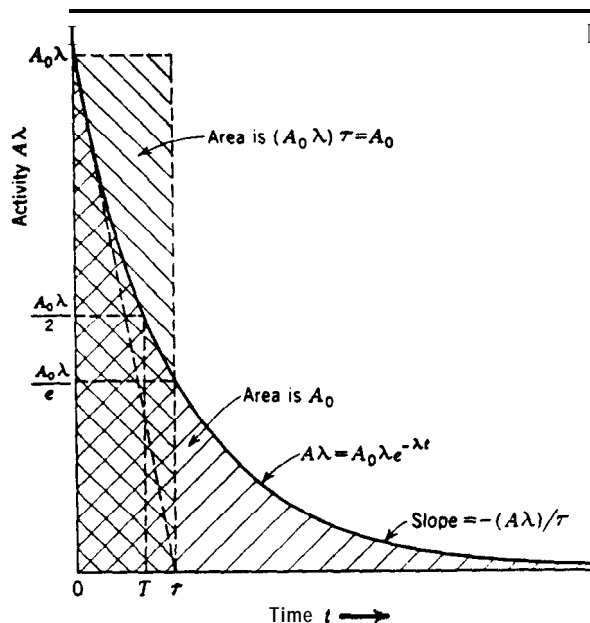


Fig. 1.1 Graphical relationships in the decay of a single radioactive nuclide. $T = 0.693/\lambda = 0.693\tau$; $\tau = 1.44T$; $\tau = 1/\lambda$. T = half-period; τ = mean life; λ = decay constant; A_0 = number of atoms at time $t = 0$; A = number of atoms at t . The slope of the activity curve at any time t is $d(A\lambda)/dt = -(A_0\lambda)\lambda e^{-\lambda t} = -\lambda(A\lambda) = -(A\lambda)/\tau$. The initial slope, at $t = 0$, is $-\lambda(A_0\lambda) = -(A_0\lambda)/\tau$. If the initial slope is extrapolated (dotted line), it intersects the time axis at the mean life τ .

vs. time equals the total number of atoms which were present initially; thus,

$$\text{Area} = \int_0^{\infty} A\lambda dt = \int_0^{\infty} \lambda A_0 e^{-\lambda t} dt = A_0 \quad (1.16)$$

This area is the same as that within the rectangle $A_0\lambda\tau$. Thus, if the initial activity $A_0\lambda$ could remain constant for a mean life τ , all the atoms would have been transformed.

g. Aids in the Computation of Radioactive Decay. Computations of $e^{-\lambda t}$ can be tedious if a few simple aids are ignored. Some of the most useful elementary techniques (K24) follow.

Semilogarithmic Graph Paper. If relative activities A/A_0 or relative amounts A/A_0 are plotted on the logarithmic axis, against time t on the linear axis, then $e^{-\lambda t}$ is a straight line passing through the points $A/A_0 = 1$ at $t = 0$ and $A/A_0 = 0.5$ at $t = T$. Depending on the scale of the graph paper, two- or three-place accuracy is obtainable, as illustrated in Fig. 12.1b.

Common (Base 10) Logarithm Tables, or Slide Rules. It may be noted that Eq. (1.3) or (1.4) can be expressed equally well using the base 10, instead of the base e . Thus

$$\frac{A_0}{A} = e^{\lambda t} = e^{\lambda T(t/T)} = 2^{(t/T)} \quad (1.17)$$

$$\ln \left(\frac{A_0}{A} \right) = \left(\frac{t}{T} \right) \ln 2 = \frac{0.693t}{T} \quad (1.18)$$

$$\log_{10} \left(\frac{A_0}{A} \right) = \left(\frac{t}{T} \right) \log_{10} 2 = \frac{0.301t}{T} \quad (1.19)$$

$$\text{or} \quad \frac{A}{A_0} = e^{-\lambda t} = \left(\frac{1}{2} \right)^{t/T} = e^{-0.693(t/T)} = 10^{-0.301(t/T)} \quad (1.20)$$

Thus, on slide rules, the value of the mantissa of $0.301(t/T)$ can be set on the L scale, and A_0/A can be read on the C scale, or A/A_0 read directly on the CI scale.

Log-Log Slide Rules. These rules have scales of e^x and of e^{-x} . At the risk of being gratuitous, it should be said that the student will be amply rewarded by becoming familiar with their range and uses. When 0.5 on the log-log scale is set opposite the half-period T , A/A_0 can be read directly for all other values of t . Analytically, the operation of the exponential scales can be illustrated by the following expressions

$$\begin{aligned} e^{-\lambda t} &= \frac{1}{2}^{t/T} & \log e^{-\lambda t} &= \frac{t}{T} \log \left(\frac{1}{2} \right) \\ \log \log e^{-\lambda t} &= \log \log \left(\frac{1}{2} \right) + \log \left(\frac{t}{T} \right) \end{aligned} \quad (1.21)$$

Approximate Forms. For values of t which are small compared with the half-period, use of the exponential expansion

$$e^{-\lambda t} = e^{-(t/\tau)} = 1 - (t/\tau) + \frac{(t/\tau)^2}{2} - \dots \quad (1.22)$$

is often the most accurate procedure.

Problems

1. The relative intensities of negatron β decay, electron-capture decay, and positron β decay of Cu^{64} are approximately (β^-) : (EC): $(\beta^+) = 2.0:2.0:1.0$. The half-period of Cu^{64} is 12.8 hr.

(a) Calculate the total decay constant λ and the partial decay constants λ_{β^-} , λ_{EC} , λ_{β^+} in sec^{-1} .