KINETICS OF RADIOACTIVE DECAY & GROWTH

\[ N(t) = N_0 e^{-\lambda t} \]
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To the Reader

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Törökbálint, July 18, 2009
Sándor Nagy, Author

Introduction

At present there are over 3000 known nuclides, 265 of which are stable, while the rest, i.e. more than 90% of them, are radioactive. The chemical applications of the specific isotopes of chemical elements are mostly connected with the latter group, including quite a number of metastable nuclear isomers, making the kinetics of radioactive decay an important chapter of nuclear chemistry. After giving a phenomenological and then a statistical interpretation of the exponential law, we discuss the various combinations of individual decay processes as well as the cases of equilibrium and non-equilibrium. Half-life systematics of the different decay modes are also summarized.

1. THE EXPONENTIAL LAW OF DECAY

1.1. Phenomenological approach

The most fundamental quantity of radioactive decay is the activity \( A \) meaning the number of atoms decaying in the specimen per time.

Historical units of activity. The formerly used unit of radioactivity—the curie (Ci)—was introduced in 1930. One curie was originally defined as the disintegration rate of 1 gram of

\[ A = \frac{dN}{dt} \]

\[ \text{Ci} = \text{disintegrations per second per gram} \]
pure $^{226}$Ra. Radium was chosen as a reference because of its then importance among radioactive elements. However it was rather disturbing that the value of the unit had to be changed from time to time as more accurate data were obtained for the decay of radium. Finally the value of curie was fixed arbitrarily at $3.7 \times 10^{10}$ disintegrations per second (1950). (During the thirties another unit: $10^6$ disintegrations per second—called rutherford—was also suggested but it has never become widely known.)

**SI unit of activity.** Since 1975, the internationally recommended unit is the becquerel (Bq) defined as 1 disintegration per second (1 dps), but workers in nuclear and radiochemistry, especially those handling high activities, still stick to the old unit curie. Some authors prefer the notation dps to Bq and the acronym dpm (meaning decays per minute) also occurs as a unit of activity.

**Intensive quantities related to the activity.** In most samples the radionuclides are not carrier-free but inactive isotopes ‘carry’ them and/or an inactive substance called matrix contains them. (The term no-carrier-added or n.c.a. only means that the production process of a radionuclide sample does not involve adding carrier to it. However it may not be carrier-free.) The **specific activity** of a radioactive sample is defined as the (absolute) activity of the radioactive sample divided by its mass (unit: Bq/kg). Similar quantities are the **molar activity** (unit: Bq/mol) and the **activity concentration** (unit: Bq/dm$^3$), defined as the activity of the sample divided by its molar amount and volume, respectively.

Note that the term specific activity is also used in the sense activity caused by a radioisotope of an element in a sample per the total mass of the element (or a compound of it) contained by the sample. Some authors use the term ‘specific radioactivity’ in the sense of molar activity, although according to IUPAP and IUPAC recommendations the adjective ‘specific’ should be reserved for naming quantities that are divided by mass. So the use of the term in the latter sense should be avoided.

**Basic equations of activity.** The activity $A$, being the measure of **disintegration rate**, is related to the number of radioactive atoms $N$ through the following expression:

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

where $\Delta t$ is the observation period and $|\Delta N|$ is the number of atoms that have decayed during $\Delta t$.

We have utilized above that if $\Delta t$ as well as $|\Delta N| / N$ are small enough—i.e., if we observe a large number of atoms in a short time interval—then $N(t)$ can be considered as a ‘smooth’ enough function for which the ratio $\Delta N/\Delta t$ can be replaced by the derivative $dN/dr$.

Radioactive decay usually produces radiation particles ($\alpha$, $\beta$) or photons ($\gamma$) that can be detected with some probability. Under favorable conditions, the observed activity $A'$ thus determined will be proportional to the actual activity (also called: **absolute activity**) $A$:

$$A' = \eta A, \quad (2)$$

where $\eta (\leq 1)$ is the efficiency of detection.

It is also an empirical fact that if we have a larger number of atoms (e.g., by preparing a larger sample from the same homogeneous radioactive material), then the activity will be proportionally larger. For a given type of radionuclide we can therefore write

$$A = \lambda N, \quad (3)$$

where the factor $\lambda$ (dimension: 1/time; unit: s$^{-1}$) is called the **decay constant**.
Note that according to Eq. (3) the quantity of a radionuclide can be specified by its activity. (This is the ‘theoretical’ background of the common practice that radioactive sources are sold/bought by the megabecquerel or millicurie.)

**Determination of the decay constant.** The above equation, combined with Eq. (2), also serves as a basis for the determination of the decay constant of long-lived radionuclides having very **long half-lives** [see Eq. (17)], which means that neither their activity nor their number is expected to change appreciably within a reasonable period of time that can be spent for their observation:

\[
\lambda = \frac{A}{N} = \frac{A'}{\eta N}.
\]  

Comparing Eqs. (1) and (3) we get

\[
\frac{dN}{dt} = -\lambda N,
\]  

which is essentially the same differential equation that is characteristic of first-order reactions in chemistry with \( \lambda \) playing the role of the rate constant.

The solution of Eq. (5) yields the formula generally referred to as the **exponential law** of radioactive decay:

\[
N = N(0) e^{-\lambda t}, \tag{6}
\]

where \( N(0) \) is the number of radioactive atoms present at \( t = 0 \).

Multiplying Eq. (6) by \( \lambda \) and then by \( \eta \), and comparing the results with Eqs. (3) and (2) we find that the same type of exponential law holds for the activities as well:

\[
A = A(0) e^{-\lambda t}, \tag{7}
\]

\[
A' = A'(0) e^{-\lambda t}. \tag{8}
\]

Note that Eq. (8) is especially useful for the determination of the decay constant of radionuclides with **medium half-lives**, i.e., whenever it is possible to ‘sit out’ the change of activity, because observed activities are a lot easier to measure than either (absolute) activities or numbers of atoms (of specific nuclides). The latter is unavoidable, e.g., in the case of long-lived radionuclides when we can only use Eq. (4) for the determination of \( \lambda \).

In the case of extremely **short half-lives**, the starting point of decay-constant determination is still something like Eq. (8), but special tricks/equipment (coincidence circuit, time-to-amplitude converter, multichannel analyzer) as well as special conditions (available start and stop signals informing of the ‘birth’ and ‘death’, respectively, of individual nuclei/particles) are also needed. This is how positron life-time distributions are determined, for instance.

In olden times the lifetime distribution of short-lived nuclides was measured without a multichannel analyzer using the **method of delayed coincidences**. In this case the start signals were delayed and the coincidence rate was plotted against the time of delay. The resulting graph showed the characteristic features of the exponential law, from which the decay constant could be determined by calculating the slope of the semilogarithmic plot.

In the case of **man-made chemical elements** (e.g. transactinides) a different type of problem arises, namely, the number of decays that can be observed is just not enough to measure out the exponential law because only a couple of like atoms are produced at a time. In such a case, the determination of the decay constant—or, more directly, that of the **mean-life**
[see Eq. (18)]—can be performed by finding the maximum of the logarithm of life-time histogram. The maximum is supposed to be at \( \log \tau \).

There is another problem with man-made chemical elements, namely that their isotopes are short lived too. This, together with the need for their identification, calls for separation methods that are very fast. Some types of equipment are able to handle radionuclides with half-lives of \( \sim 10 \) s or even \( \sim 1 \) s.

### 1.2. Stochastic approach

In the text ‘Stochastics and Nuclear Measurements’, we have discussed the connection between the exponential law and some of the special distributions—i.e. binomial, Poisson, and exponential—in more detail. Here we only wish to give a simple illustration of the stochastic features.

Consider a single radioactive atom of some kind. Let \( p \) denote the probability that it will disintegrate within a certain observation period \( \Delta t \). Let us assume that the atom is ageless in the sense that \( p \) is independent of how long it had existed before we started its observation. It stands to reason that the chance of decay increases with the length of the observation period. Moreover, it also seems reasonable to assume that for short enough \( \Delta t \) we have proportionality

\[
p = \lambda \Delta t ,
\]

(9)

where the proportionality constant \( \lambda \) only depends on the type of the atom (or, more specifically, on its readiness to decay).

However we are more interested in the complementary event, i.e., that the atom is still intact after the period \( \Delta t \). The probability of this is:

\[
P(\Delta t) = 1 - p = 1 - \lambda \Delta t .
\]

(10)

Suppose that the atom has indeed survived the first \( \Delta t \) period of observation. Then the probability of its being ‘alive’ even after the next \( \Delta t \) period is again

\[
1 - p = 1 - \lambda \Delta t .
\]

According to probability theory, for the total observation time \( 2 \Delta t \) we have

\[
P(2 \Delta t) = (1 - \lambda \Delta t)^2 .
\]

(11)

Thus the probability that the atom will survive a period of \( n \Delta t \) is

\[
P(n \Delta t) = (1 - \lambda \Delta t)^n
\]

(12)

which, with the notation \( n \Delta t = t \), can be rewritten as

\[
P(t) = \left(1 - \frac{t}{\lambda n}\right)^n .
\]

(13)

For \( n \to \infty \) (i.e., when the division of \( t \) to single periods of observation is infinitely fine) we get the following expression for the probability that the atom does not decay in an arbitrarily long period \( t \):

\[
P(t) = e^{-\lambda t} .
\]

(14)

For a large number of identical atoms, the exponential factor in Eq. (14) represents the fraction of survivors \( N(t)/N(0) \) as a function of the time \( t \). Thus the initial number of atoms \( N(0) \) is expected to drop to \( N(t) \) according to the following formula:
\[ N(t) = N(0) e^{-\lambda t}. \]  

(15)

Note that what we have just ‘derived’ is the **exponential law** of decay [see Eq. (6)] justifying the notation \( \lambda \) (reserved for the decay constant throughout this chapter) for the proportionality constant in Eq. (9).

Using Eq. (15), we can introduce a more expressive quantity than the decay constant for the description of the ‘decayability’ of radionuclides. The **half-life** \( T_{1/2} \) is the time in which half of the atoms are expected to decay:

\[ N(T_{1/2}) = N(0) e^{-\lambda T_{1/2}} = \frac{N(0)}{2}, \]

whence

\[ T_{1/2} = \frac{\ln 2}{\lambda} \approx 0.693 \frac{\lambda}{\lambda}. \]  

(17)

Another characteristic time parameter of radioactive decay is the **mean life** \( \tau \), which is the expected value of the \( \gamma(1, \lambda) \) exponential distribution associated with radioactive decay:

\[ \tau = \int_0^\infty t f(t) \, dt = \lambda \int_0^\infty te^{-\lambda t} \, dt = \frac{1}{\lambda}, \]

(18)

where \( f(t) = \lambda e^{-\lambda t} \) is the density function of the **lifetime distribution**.

Comparing this result with Eq. (17), we find that the mean life is proportional to the half-life:

\[ \tau = \frac{T_{1/2}}{\ln 2} \approx 1.44 T_{1/2}. \]  

(19)

It is common knowledge that \( \tau \) is the time necessary for the atoms to be reduced to \( 1/e \) of their initial number:

\[ N(\tau) = N(0) e^{-\frac{1}{\lambda}} = \frac{N(0)}{e} \approx 0.37 N(0). \]  

(20)

We can see that the drop in the number of survivors is larger than in the case of the half-life. The reason is obvious: according to Eq. (19) \( \tau > T_{1/2} \).

Note that the ‘\( 1/e \) rule’ expressed by Eq. (20) is not a general criterion for mean times: it is a specific feature of the exponential distribution associated with the exponential law of simple decays. (See also the comment after Eq. (44).)
FIGURE 1. Schematic representation of the logarithm of activity for a source containing the mixture of two radionuclides. We can see that nuclide 1, due to its longer mean life, will survive nuclide 2, although its initial activity at $t = 0$ is much lower (10% of the total). This fact used to be the basis of the graphical decomposition of the decay curves of mixtures in the ‘pre-PC’ age of nuclear science. The idea for obtaining the $A_2(t)$ values (red line) from the thick (black) curve of the experimental $A(t)$ values was to subtract the values of $A_1(t)$ extrapolated back along the blue line representing the asymptote of the thick line for $t \to \infty$.

2. INDEPENDENT DECAYS—RADIONUCLIDE MIXTURES

If we find that the logarithmic plot of activity versus time is not a straight line, then this is an indication that the sample contains more than one radionuclide. The radionuclides thus detected may be genetically related to each other (i.e. one is produced by the other through radioactive transformation), or they can be independent of each other, forming a simple mixture (see FIGURE 1). In this section we are going to deal with the latter case.

In order to determine the individual decay constants of the mixture, we must measure count rates (i.e. observed activities) as a function of time, provided that the half-lives are moderately long and comparable with each other. The number of measured points should be at least twice the number of radioactive components plus one (or, preferably, a lot more).

The analysis of the ‘nuclear spectrum’ thus obtained is based on the fact that the observed activities are additive and also that they are subject to the exponential law expressed by Eq. (8). Hence, for $n$ radioactive components we have

$$A'(t) = \sum_{i=1}^{n} A'_i(t) + b = \sum_{i=1}^{n} A'_i(0) e^{-\lambda_i t} + b$$

(21)

where $A'(t)$ is the observed activity of the mixture at time $t$, $A'_i(0)$ is the observed activity of the $i$th component at $t = 0$ and $b$ is the background. The parameters $(2n + 1$ altogether), including the decay constants, can then be estimated by a least-square fit.
3. BRANCHING DECAYS

Some radionuclides can disintegrate in two or more competing ways leading to different products. In the case of odd-odd nuclei, e.g., sitting nearest to the apex of one of the mass parabolas, negative β decay and electron capture (and in energetically favorable cases: positive β decay) are equally possible. For instance, the nucleus of $^{40}$K, a nuclide used in radioactive dating, can undergo both electron capture and β− decay (see Figure 2 for more detail):

$$\lambda = \lambda_{EC} + \lambda_{\beta^{-}}, \quad ^{40}K \xrightarrow{^\lambda} ^{18}Ar, \quad ^{40}K \xrightarrow{^\lambda} ^{20}Ca.$$  \hspace{1cm} (22)

In the case of the decay series shown by Figures 4-7, another type of branching decay occurs: α decay competes with β− decay. For instance, $^{227}$Ac, the eponymous member of the actinium (4n+3) series (see Figure 7), undergoes the following disintegration:

$$\lambda = \lambda_{EC} + \lambda_{\beta^{-}}, \quad ^{227}Ac \xrightarrow{^\lambda} ^{87}Fr, \quad ^{227}Ac \xrightarrow{^\lambda} ^{90}Th.$$  \hspace{1cm} (23)

The additive property of the partial decay constants indicated above follows from the same property of the respective partial decay rates. In the case of $^{40}$K, e.g., we can write

$$-\frac{dN_K}{dt} = \frac{dN_{Ar}}{dt} + \frac{dN_{Ca}}{dt} = \lambda_{EC}N_K + \lambda_{\beta^{-}}N_K = (\lambda_{EC} + \lambda_{\beta^{-}})N_K = \lambda N_K.$$  \hspace{1cm} (24)

![Figure 2](image-url)  
**Figure 2.** Mass parabolas explaining the branching decay of $^{40}$K. This type of branching is always associated with odd-odd nuclei. The parabolic dependence of the nuclear mass on the atomic number $Z$ comes from a similar (but negative) dependence of the binding energy as expressed by the Weizsäcker formula. (The odd-odd parabola and the even-even one are shifted relative to each other by $2\delta/A^{3/4}$ according to the Weizsäcker formula. Note that in the case of odd-$A$ isobars there is only one mass parabola (and therefore one minimum) because the pairing energy term of the Weizsäcker formula is the same (zero) for even-odd and odd-even nuclei.) Note that $^{40}$Ca is a doubly magic nuclide, which explains why it has a lower mass (higher stability) than expected from the general trend expressed by the lower (blue) parabola.
FIGURE 3. Schematic representation of branching decay for $\lambda_1/\lambda_2 = 3$. The arrows shown between the curves of the daughters have the same length as the one displayed at the vertical axis between 1 and 3. The constant separation—considering that the vertical scale is logarithmic—means that the upper curve, representing daughter 1, is a scaled-up version of the lower curve. The value of the scaling factor (3) is determined by the ratio $\lambda_1/\lambda_2$. [See Eq. (30).]

Hence the half-life of $^{40}$K according to Eq. (17) is

$$T_{1/2} = \frac{\ln 2}{\lambda} = \frac{\ln 2}{\lambda_{EC} + \lambda_{\beta}}.$$  \hspace{1cm} (25)

According to Eqs. (24) and (6) the growth rates of $^{40}$Ar and $^{40}$Ca are

$$\frac{dN_{Ar}}{dt} = \lambda_{EC} N_K = \lambda_{EC} N_K(0) e^{-\lambda t},$$  \hspace{1cm} (26)

$$\frac{dN_{Ca}}{dt} = \lambda_{\beta} N_K = \lambda_{\beta} N_K(0) e^{-\lambda t}.$$  \hspace{1cm} (27)

The numbers of the respective nuclides that are present at time $t$ can be calculated by integration. If we assume that at $t = 0$ neither $^{40}$Ar, nor $^{40}$Ca was present, then we get

$$N_{Ar} = \lambda_{EC} \frac{N_K(0) (1 - e^{-\lambda t})}{\lambda},$$  \hspace{1cm} (28)

$$N_{Ca} = \lambda_{\beta} \frac{N_K(0) (1 - e^{-\lambda t})}{\lambda}.$$  \hspace{1cm} (29)

Thus we find that the atomic ratio of $^{40}$Ar to $^{40}$Ca is equal to the ratio of the partial decay constants called branching ratio at any moment:

$$\frac{N_{Ar}}{N_{Ca}} = \frac{\lambda_{EC}}{\lambda_{\beta}}.$$  \hspace{1cm} (30)
Therefore the atomic fraction of either product is indicative of the contribution of the respective partial decay constant to the total \( \lambda \). For instance, in the case of \(^{40}\)Ar produced by EC we obtain

\[
x_{\text{Ar}} = \frac{N_{\text{Ar}}}{N_{\text{Ar}} + N_{\text{Ca}}} = \frac{\lambda_{\text{EC}}}{\lambda_{\text{EC}} + \lambda_{\beta}} = \frac{\lambda_{\text{EC}}}{\lambda}.
\]

Hence, for the products of several parallel decays we can write

\[
\lambda_j = x_j \sum_{k=1} \lambda_k = x_j \lambda.
\]

4. SUCCESSIVE PROCESSES

4.1. General kinetics of decay chains

It often happens that the decay product itself is also radioactive. Thus a whole chain of radioactive transformations may evolve, forming a radioactive decay series [see FIGURES 4-7].

Most of the naturally occurring radionuclides belong to one of three series shown in FIGURES 4, 6, and 7 where the symbols of the elements that are represented in the given series are put in boxes. The primary parent and the final product of each series are indicated by shading the box of the respective elements.

It can be useful to determine the activity of a given member of a series at a given time \( t \). Let us deal with the simplest case first, i.e., when the first two members (the parent \( X_1 \) and its daughter \( X_2 \)) are only considered:

\[
X_1 \xrightarrow{\lambda_1} X_2 \xrightarrow{\lambda_2} \rightarrow
\]

\[
N_1 \xrightarrow{\lambda_1} N_2
\]

where \( N_1 \) and \( N_2 \) are the numbers of atoms of \( X_1 \) and \( X_2 \), respectively. (Some authors prefer to use the term mother rather than parent.)

The time dependence of the number of atoms and the activity (decay rate) of the parent \( X_1 \) follows directly from Eqs. (6) and (7):

\[
N_1 = N_1(0) \, e^{-\lambda_1 t},
\]

\[
A_1 = -\frac{dN_1}{dt} = \lambda_1 N_1 = \lambda_1 N_1(0) e^{-\lambda_1 t} = A_1(0) e^{-\lambda_1 t}.
\]

The daughter \( X_2 \) forms at a rate equal to the decay rate of the parent and simultaneously disintegrates at a rate determined by \( \lambda_2 \):

\[
\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2.
\]

Substituting Eq. (34) into Eq. (36) we get

\[
\frac{dN_2}{dt} + \lambda_2 N_2 = \lambda_1 N_1(0) e^{-\lambda_1 t}.
\]
The solution of Eq. (37) for the initial conditions $t = 0$: $N_1 = N_1(0), N_2 = 0$ is

$$N_2 = N_1(0) \frac{\lambda_1}{\lambda_2 - \lambda_1} \left( e^{-\lambda_1 t} - e^{-\lambda_2 t} \right).$$

(38)

Thus the activity of the daughter is

$$A_2 = \lambda_2 N_2 = A_1(0) \frac{\lambda_2}{\lambda_2 - \lambda_1} \left( e^{-\lambda_1 t} - e^{-\lambda_2 t} \right).$$

(39)

**FIGURE 4.** The thorium ($4n + 0$) series is one of the four decay series, out of which three (including this one) are naturally occurring, while one (the $4n+1$ series) is extinct on Earth. The numbering—$4n+0$, $4n+1$, $4n+2$, and $4n+3$—refer to the residue class (modulo 4) of the possible mass numbers in each series. The explanation for the notation is that these series (see also Figures 5-7 showing the same range of $Z$ and $A$ for better comparison) only include $\alpha$ decay (slanting arrows) and $\beta$ decay (horizontal arrows), so the change of the mass number is either 4 or 0. Therefore, the members of the whole series belong to the same residue class (modulo 4) as their primary parent. (There are also some very weak branches of decay indicated in parentheses like spontaneous fission SF and different types of cluster decay when a C, O or even a Ne nucleus is emitted instead of an $\alpha$ particle.) Each of the four series represents an example for secular equilibrium due to the very long half-life of the primary parent of the series in comparison with any of the members. For instance, the half-life of $^{232}$Th is $T_{1/2} = 1.405 \times 10^{10}$ a—a value close to the estimated age of the Universe—while that of the longest-lived daughter, $^{228}$Ra, is only $T_{1/2} = 5.75$ a.

More generally, the following system of differential equations is to be solved:

$$\frac{dN_1}{dt} = -\lambda_1 N_1,$$

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2,$$
\[ \frac{dN_1}{dt} = \lambda_2 N_2 - \lambda_3 N_3, \]

\[ \vdots \]

\[ \frac{dN_n}{dt} = \lambda_{n-1} N_{n-1} - \lambda_n N_n. \]  

(40)

FIGURE 5. The neptunium \((4n+1)\) series is now extinct on Earth. This is due to the relatively short half-life of the primary parent \(^{237}\)Np \((T_{1/2} = 2.14 \times 10^6 \text{a})\), which is less than the age of the human race. The nearly 2136 half-lives, that have elapsed since the creation of our planet (about \(4.57 \times 10^9 \text{ years ago}\)), have practically wiped out this process because only 1 out of \(10^{643}\) neptunium-237 atoms is expected to survive. Now, the odds of survival are really bad, considering that \(10^{643}\) Np atoms would ‘weigh’ about \(3 \times 10^{618} \text{ kg}\), whereas the mass of the Sun is a mere \(2 \times 10^{30} \text{ kg}\), and even that of the Milky Way is only estimated to about \(2 \times 10^{41} \text{ kg}\).

The initial conditions considered are

for \(t = 0\): \(N_1 = N_1(0); N_2 = N_3 = \ldots N_n = 0. \)  

(41)

The solution of the above system of equations can be found, e.g., in ref. Vértoves and Kiss 1987. [Ehmann and Vance (1991) as well as several authors of this handbook refer to the result summarized by Eqs. (42)-(43) as the Bateman equation (Bateman 1910). Other authors refer to the whole series of equations shown in Eq. (40) as Bateman equations.] For the \(n\)th member of the series \((n = 1, 2, 3\ldots)\) we get

\[ N_n = N_1(0) \frac{f_n(t)}{\lambda_n} \equiv N_1(0) \sum_{k=1}^{n} a_{kn} e^{-\lambda_k t} \]  

(42)

where \(a_{11} = 1\), and the rest of the summation coefficients \(a_{kn}\) are given by the expression
FIGURE 6. The uranium \( (4n + 2) \) series is the second of the three naturally occurring decay series on Earth. The half-life of the primary parent \( ^{238}\text{U} \) \( (T_{1/2} = 4.5 \times 10^9 \text{ a}) \) nearly matches the estimated age of our planet, so about half of the \( ^{238}\text{U} \) atoms that once existed are still around. In this series Mg emission also occurs some of the \( ^{210}\text{Pb} \) nuclei undergo beta-delayed neutron emission. This establishes a vague interconnection with the \( (4n+1) \) series.

The above Bateman equations have also been generalized to include activation by neutron capture.

For all the sophistication of the above result, some questions are easy to answer without using the explicit formulae. Such a question is, e.g., the mean time needed for the parent nucleus (X1) to go through \( n \) steps of subsequent transformations. (For instance, we may be interested in the mean time needed for a \( ^{238}\text{U} \) atom to transform to \( ^{206}\text{Pb} \) through the series of decays shown in Figure 6.) Instead of integrating Eq. (42) (or, actually, \( t_{f_{kn}}(t) \)), we will translate the problem to a probability problem in the following way.
FIGURE 7. The actinium \((4n+3)\) series is the third one of the naturally occurring decay series on Earth. The half-life of the primary parent \(^{235}\text{U}\) \((T_{1/2} = 7.04 \times 10^8 \text{ a})\) — note, for comparison, that the Cambrian Period started some \(5.7 \times 10^8\) years ago) is approximately one sixth of the estimated age of the Earth. Thus, nearly \(2^{-6}\) part (or, more exactly, just about 1\%) of the \(^{235}\text{U}\) atoms that once existed are still potentially available as nuclear fuel.

The subsequent periods of time \(T_i\) that are spent by a nucleus at the \(i\)th ‘stage’ \((i = 1, 2, \ldots, n)\) are random variables with \(\gamma(1, \lambda_i)\) exponential distributions. The total waiting time to get through \(n\) stages is \(T = T_1 + \ldots + T_n\). (Note that according to Rényi 1979 the function \(f_\nu(t)\) in Eq. (42) plays the role of the density function of \(T\).) What we actually need is the expected value \(\tau\) of \(T\). Making use of the fact that expected value of the sum of random variables always equals the sum of the expected values, we can directly write up the following result:

\[
\tau = \sum_{i=1}^{n} \tau_i = \sum_{i=1}^{n} \frac{1}{\lambda_i}.
\]

(44)

Note that the ‘1/e rule’ [see Eq. (20)] does not apply to \(\tau\), because the distribution of \(T\) is only exponential in the trivial case of \(n = 1\). To give a numerical example, let us take the case of nearly equal decay constants. If \(\lambda_1 \approx \lambda_2 \approx \lambda\), then \(T\) has a \(\gamma(n, \lambda)\) gamma distribution for which \(\tau = n/\lambda\). One can easily verify, e.g. for \(n = 2\), that the factor in question would then be \(1 - F_2(2/\lambda) = 3e^{-2} \approx 0.41\) rather than \(e^{-1} \approx 0.37\), where \(F_2(t)\) is the distribution function of the \(\gamma(2, \lambda)\) gamma distribution. In the given example, this would mean that about 59\% \((= 100\% - 41\%)\) of the initial atoms rather than 63\% \((= 100\% - 37\%)\) are expected to undergo both decays by the time \(t = \tau = 2/\lambda\). For large values of \(n\), on the other hand, we get 50\%, because the gamma distribution can be approximated by normal distribution for which the expected value \(\tau\) and the median \((T_{1/2})\) are equal.

In one particular (however important) type of radioactive series, we still find the ‘1/e rule’ working. This happens when the half-life (and therefore the mean life) of one of the members is overwhelmingly large compared with all the others. This case, called secular equilibrium, is discussed separately due to its importance.
4.2. Radioactive equilibrium and non-equilibrium

It is often not necessary to bother with the general solution of the successive decay problem, because after some time the whole series can be analyzed stepwise, comparing the decay constants of the individual decays. In order to introduce the concepts of equilibrium and non-equilibrium, we start with the analysis of the first two decays of the series shown by Eq. (33). Then, we will include the decay of the second daughter as well, in order to be able to draw more general conclusions concerning the whole of the series. Under certain conditions some kind of equilibrium is established between the (primary) parent and its (first) daughter.

The condition for transient equilibrium—or any equilibrium for that matter—is: \( \lambda_1 < \lambda_2 \) (or \( \tau_1 > \tau_2 \)). In this case, after some time we have

\[
e^{-\lambda_1 t} \gg e^{-\lambda_2 t},
\]

and therefore the second term becomes negligible in Eqs. (38) and (39).

Thus, the asymptotic behavior of the number of the daughter’s atoms \( N_2 \) is described by the following formula:

\[
N_2 \Rightarrow \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1(0) e^{-\lambda_1 t} = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1.
\]

(46)

From Eq. (46) we get for the asymptotic ratio of the numbers of atoms

\[
\frac{N_2}{N_1} \Rightarrow \frac{\lambda_1}{\lambda_2 - \lambda_1},
\]

(47)

which shows that, for large enough \( t \), the ratio of the number of daughter atoms to that of the parent atoms \( (N_2/N_1) \) becomes approximately constant.

The asymptotic behavior of the daughter’s activity \( A_2 \) is obtained from Eq. (39) using Eq. (45):

\[
A_2 \Rightarrow \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1(0) e^{-\lambda_1 t} = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1.
\]

(48)
FIGURE 8. Schematic representation of (transient) equilibrium. The half-life of the parent nuclide considered is 10 units, while that of the daughter is 1 unit. Note that the maximum of the daughter’s activity [see Eq. (58)] is reached somewhat later than that of the total activity. Note also that Eq. (53) works quite well, i.e., the logarithmic curves of $A_1$ and $A_2$ are already practically parallel at $t \approx 6.6$, which means that the ratio $A_2/A_1$ is nearly constant.

Thus we obtain for the asymptotic ratio of the activities

$$\frac{A_2}{A_1} \Rightarrow \frac{\lambda_2}{\lambda_2 - \lambda_1}.$$  \hfill (49)

We can see that the ratio of the activities also becomes constant after some time. This can also be observed in FIGURE 8, where proportionality reveals itself as a constant separation between the logarithmic curves of $A_1$ and $A_2$.

It can also be concluded from Eq. (49) that in transient equilibrium the daughter is more active than the parent. (We can also see this in FIGURE 8.)

We can set up the following criterion for the minimum time required to approach the equilibrium condition defined by Eq. (49) to a given relative accuracy $\alpha$:

$$\frac{\lambda_2}{\lambda_2 - \lambda_1} - \frac{A_2}{A_1} \leq \alpha,$$

$$\frac{\lambda_2}{\lambda_2 - \lambda_1} - \frac{A_2}{A_1} \leq \alpha,$$

where $A_2/A_1$ is the actual ratio of the activities as a function of time, i.e.,

$$\frac{A_2}{A_1} = \frac{\lambda_2}{\lambda_2 - \lambda_1} (1 - e^{-(\lambda_2 - \lambda_1)t})$$  \hfill (51)

as follows from Eqs. (39) and (35).

Hence, criterion (50) can be rewritten in the following form:
\[ e^{-(\lambda_2 - \lambda_1) t} \leq \alpha, \]  
which eventually yields

\[ t > \tau_2 \ln \frac{1}{\alpha}, \]  

where \( \tau_2 \) is the mean life of the (shorter-lived) daughter nuclide.

If, e.g., we set a criterion of 1%, then we get from Eq. (53): \( t > 4.6 \tau_2 \approx 6.6 T_{1/2} \). In other words, after about 6-7 half-lives (4-5 mean lives) of the daughter nuclide we get about 1% close to the equilibrium condition.

The condition for **secular equilibrium** is: \( \lambda_1 << \lambda_2 \) (or \( \tau_1 >> \tau_2 \)). In this case \( \lambda_1 \) can be neglected in comparison with \( \lambda_2 \) and Eq. (49) becomes even simpler:

\[ \frac{A_2}{A_1} \rightarrow 1, \]  

that is, in secular equilibrium we have

\[ A_1 \approx A_2. \]

Secular equilibrium is illustrated by FIGURE 9 in the special case of \(^{226}\text{Ra}\) as the parent \((T_{1/2} = 1600 \text{ a } = 584 \text{ 000 d})\) and \(^{222}\text{Rn}\) as its daughter \((T_{1/2} = 3.82 \text{ d})\).

If \( \lambda_1 > \lambda_2 \), then **no equilibrium** is reached at any time. Still, similarly to the case of equilibrium, the activity of the daughter passes through a maximum (see FIGURE 10).

The **asymptotic behavior of the daughter's activity** \( A_2 \) is now different from that shown by Eq. (48), because it is the first term that becomes negligible in Eq. (39):

\[ A_2 \Rightarrow \frac{\lambda_2}{\lambda_1 - \lambda_2} A_1(0) e^{-\lambda_2 t}. \]  

Note that the above equation would describe the activity of a source prepared from the daughter's atoms, with the initial activity

\[ A_2(0) = \frac{\lambda_2}{\lambda_1 - \lambda_2} A_1(0). \]  

(See the intersection of the dotted line with the vertical axis in FIGURE 10.)
FIGURE 9. The change of activity in the case of secular equilibrium. The half-life of the parent nuclide $^{226}\text{Ra}$ is $T_{1/2} = 1600$ a, and that of the daughter $^{222}\text{Rn}$ is $T_{1/2} = 3.825$ d. The time unit is the half-life of the daughter. Note that after the equilibrium has been reached, the activities of Ra and Rn become practically equal—the distinguishing feature of secular equilibrium. This means that the total activity doubles after 6-8 half-lives of the daughter.

It can be useful to know the time when the daughter reaches maximum activity, especially when the aim is to collect a radionuclide produced by radioactive decay.

The maximum of $A_2(t)$ (as well as of $N_2(t)$) is reached at the time

$$t_{\text{max}} = \frac{1}{\lambda_2 - \lambda_1} \ln \frac{\lambda_2}{\lambda_1},$$

as one can easily verify by differentiating Eq. (39) (or Eq. (38)).

Let us now extend our analysis to the three first decays of the series:

$$X_1 \rightarrow X_2 \rightarrow X_3 \rightarrow \cdots$$

$N_1 \quad N_2 \quad N_3$.

Using Eqs. (42) and (43), we get for the activity of the second daughter

$$A_3 = \lambda_3 N_3 = A_1(0) \lambda_2 \lambda_3 \left( \frac{e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} + \frac{e^{-\lambda_3 t}}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} \right).$$
FIGURE 10. Schematic representation of non-equilibrium. The half-life of the parent nuclide is taken as the time unit, and that of the daughter is 10 times longer. Note that the asymptote of the daughter’s activity reflects the time characteristics of the daughter’s own, whereas in the case of equilibrium it is determined by that of the parent. (See the horizontal red line in FIGURE 9, representing both the parent’s activity and the asymptotic activity curve of the daughter.)
FIGURE 11. Basic cases of decay series consisting of 3 steps. The balanced scales draw attention to the range where the logarithms of activities become parallel, an indication of equilibrium (>). The shortest mean life is selected as the time unit for each graph. The most interesting case is represented by the bottom left graph, showing that whenever the parent of the series is the longest-lived nuclide, sooner or later equilibrium will prevail along the whole series, no matter how the individual decay constants of the daughters are related to each other. According to the four upper graphs, the longest-lived member will eventually survive (all of) its predecessor(s).

FIGURE 11 shows the activities vs. time for the three first nuclides of a series as calculated from Eqs. (35), (39) and (60). The individual graphs represent the 6 possible permutations of a fast, a medium, and a slow decay characterized by mean lives 1, 3, and 9 time units,
respectively. Careful analysis of the graphs enables us to draw general conclusions about the asymptotic behavior of any series:

If \( X_k \) is the longest-lived member of the series, then—after a sufficiently long time—the members \( X_1, \ldots, X_{k-1} \) will die out, and the series will be shortened to the sub-series \( X_k, X_{k+1}, \ldots \) with \( X_k \) as the primary parent.

No matter how the decay constants \( (\lambda_{k+1}, \lambda_{k+2}, \ldots) \) of the daughters of \( X_k \) are related to each other, the fact that \( \lambda_k \) is smaller than the rest of the decay constants enforces an equilibrium between \( X_k \) and all of its daughters.

For instance, the first member of any of the naturally occurring radioactive series has a half-life of several orders of magnitude longer than any of its descendants (see FIGURES 4-7). Therefore its decay constant \( (\lambda_1) \) is negligible in comparison with those of the decay products \( (\lambda_2, \lambda_3, \ldots, \lambda_n) \). Similarly, the exponentials

\[
e^{-\lambda_1 t}, e^{-\lambda_2 t}, \ldots, e^{-\lambda_n t}, \ldots
\]

can also be neglected in comparison with \( e^{-\lambda_1 t} \) after some time.

Everything considered, the first member of such a series is in secular equilibrium with all the others, i.e., Eq. (55) applies to any of the daughter nuclides:

\[
A_1 \approx A_2 \approx \ldots \approx A_n \approx \ldots
\]

If, therefore, all the decay products of uranium or thorium remain together in a rock or in an ancient compound, their activities become equal (after a period of time).

### 4.3. Decay after activation

A special case of successive processes is that in which the parent \( (X_1) \) is stable, but it is induced to transformation by a nuclear reaction, the product of which (the ‘daughter’ \( X_2 \)) is radioactive. For instance, in the case of neutron activation instead of Eq. (33) we can write

\[
X_1 \xrightarrow{\phi \sigma_1} X_2 \xrightarrow{\lambda_1} N_1 \rightarrow N_2,
\]

where \( \phi \) is the neutron flux density (neutrons cm\(^{-2}\) s\(^{-1}\)) and \( \sigma_1 \) is the cross section of the nuclear reaction induced by the neutrons (e.g., neutron capture):

\[
X_1(n, \gamma)X_2.
\]

Similar processes have an important part in neutron activation analysis (NAA) as well as in natural processes such as the formation of radiocarbon in the atmosphere via the reaction

\[
^{14}N(n, p)^{14}C.
\]

The equivalent of Eq. (36) is now

\[
\frac{dN_2}{dt} = \phi \sigma_1 N_1 - \lambda_2 N_2.
\]
\[ A_2 = \lambda_2 N_2 = \phi \sigma_1 N_1 (1 - e^{-\lambda_2 t}). \]  

(67)

**FIGURE 12.** Schematic representation of the activity of the radioactive product \( X_2 \) as a function of the time of activation. The time unit is the half-life of the product. The arrows point to the time calculated from Eq. (53), which now provides the minimum activation time needed to get 100\( \alpha \)% close to the asymptotic (i.e. the highest possible) value of the activity \( A_2 \Rightarrow \phi \sigma_1 N_1 \). As we can see, the gain in product activity halves in each subsequent half-life period, which makes longer irradiation times increasingly uneconomical from the viewpoint of radionuclide production.

We can see from the above equation that for a constant time of activation, the activity of the product will be proportional to \( N_1 \), offering a simple technique for the quantitative analysis of the nuclide \( X_1 \) by NAA. According to **FIGURE 12**, the asymptotic limit of the activity is also proportional to \( N_1 \). This fact is used, e.g., in the case of **radiocarbon dating**, in which the date of the death of an organism is eventually determined from the following type of equation:

\[ A_2(t) = A_2(0) e^{-\lambda_2 t} \propto \phi \sigma_1 N_1 e^{-\lambda_2 t}. \]  

(68)

5. **DECAY CONSTANT (HALF-LIFE) SYSTEMATICS**

In this section we will shortly summarize some results concerning the dependence of the decay constant on various physical parameters for the most important decay modes. The reader can find a very good theoretical introduction to this field in Friedlander et al. 1981.

5.1. **Alpha decay**

Alpha emitters are found among the large-\( Z \) nuclides (e.g., \( ^{144}_{60} \text{Nd} \) and above) for which, not too far from the bottom of the stability valley (see **FIGURE 13**), the number of neutrons exceeds that of the protons, i.e., the ratio \( N/Z \) is greater than 1. Under such conditions, the alpha decay,

\[ _Z^A X_N \rightarrow^\alpha_{Z-2} ^{4-4} X_{N-2}^2 +^4 \text{He}^2_2, \]  

(69)

will further increase this ratio
\[
\frac{N - 2}{Z - 2} > \frac{N}{Z} > 1. \tag{70}
\]

Therefore, if a nuclide is situated at the low-\(N\) side of the valley (i.e. its \(N/Z\) ratio is less than for its most stable neighbors), alpha decay can move it closer to the bottom of the valley and so the nuclide is apt to undergo alpha decay. (In certain cases even neutron-rich nuclei, e.g. some Po isotopes, may undergo \(\alpha\) decay because the \(Q\)-value of the process turns out to be positive.)

During the early decades of nuclear physics it had been thought that the alpha radiation of each alpha emitting nuclide was monoenergetic. It was not till 1929 that Rosenblum (1930) discovered the alpha radiation of \(^{212}\text{Bi}\) consisting of alpha particles of different energies. Further experiments have demonstrated that alpha energies are always discrete even in such cases, and the energy excess still possessed by the nucleus above its ground level is emitted in the form of a gamma photon.

\textbf{FIGURE 13.} The stability valley of nuclei. The vertical projection of the bottom of the valley on the \(ZA\) plane follows the line of stable nuclei at an ‘altitude’ expressed by the ratio of the nuclidic mass \((M)\) and the mass number \((A)\). The isobaric cross sections of the valley are parabolic as a consequence of the Weizsäcker formula. (See also the legend of FIGURE 2.) The local dips marked are related to magic numbers (Ca: \(N = 20\); Fe-Ni: \(N = 28\), \(Z = 28\); Y: \(N = 50\); Ba: \(N = 82\)). Beta decay occurs all along the valley (on the near side of the valley we can observe \(\beta^-\), on the far side \(\beta^+\) decay and electron capture (EC) as indicated on the parabola). Alpha decay occurs for \(A > 60\) mainly on the far side of the valley.

It should be noted that a different value of \(\lambda\) belongs to each different value of \(E_\alpha\). Thus the effective \(\lambda\) (as well as the effective \(T_{1/2}\)) for an \(\alpha\)-emitting nucleus is determined by the sum of partial decay constants; \(\sum \lambda_i\). (Note that we have a type of branching decay here.)
Alpha energies are usually between 4-6 MeV and these limits are exceeded only in a few cases. The total variation is marked by $^{144}_{60}$Nd (1.8 MeV) and $^{212}_{86}$Po (11.7 MeV). On the other hand, the half-lives cover about 30 decimal magnitudes from $^{213}_{85}$At (10$^{-7}$ s) to $^{148}_{62}$Sm (10$^{16}$ a ≈ 3×10$^{23}$ s). And we have not included the process $^{4}$Be→2$\alpha$ keeping the alpha burning in massive stars under control. The half-life of this cosmologically important decay is only about 10$^{-16}$ s or so.

There is a negative correlation between half-life and alpha energy. The strong energy-dependence of partial half-lives explains why the complexity of alpha spectra remained undetected for such a long time: the branch producing the most energetic alpha particles often dominates the whole decay process.

The first decay-constant systematics proposed is called the Geiger-Nuttal rule. This rule, set up originally as a decay-constant–alpha-range relationship, can be readily converted into the following half-life–alpha-energy relationship:

$$\log \{T_{1/2}\} = a \log \{E_\alpha\} + b,$$

(71)

where $a < 0$ is a constant and $\{x\}$ stands for the numerical value of the physical quantity $x$ in some unit $[x]$. On the other hand, $b$ has a different (but constant) value for each decay series. So the logarithmic plots of the individual series are represented by parallel straight lines shifted according to the difference in the parameter $b$.

We should note that the Geiger-Nuttal plot of the $4n+3$ series is way above the $4n+0$ and $4n+2$ series (see Figure 14). This is so because odd-Z and odd-N nuclei decay more slowly than even-even ones owing to shell effects. This kind of hindrance has saved, e.g., the very important even-odd nuclide $^{235}_{92}$U$_{143}$ — the primary parent of the $4n+3$ series — from extinction on Earth (Friedlander et al. 1981).

![Figure 14](image-url) 

**Figure 14.** Modern Geiger–Nuttal plot for the $\alpha$-decaying nuclides of the four major decay chains including the three naturally occurring ones (starting from $^{232}$Th, $^{238}$U, and $^{235}$U) once studied by the eponyms of this type of log–log presentation. The label of the fourth series (starting from $^{237}$Np) is in gray color to indicate that this series has been wiped out from Earth due to its relatively short half-life. Note that the $\alpha$ half-lives sweep the range from less than a microsecond (see the labels along the right vertical axis) to 15 billion years (the estimated age of our Universe), which means a scale of ~24 decimal or ~80 binary orders of magnitude. The $Q$-values ($-E_{\alpha}$), on the other hand, only drop from...
9 MeV to about 4 MeV, a range just about a little wider than 1 binary order of magnitude (i.e. a range covered by a factor of 2).

There are other expressions too describing such systematics.

The empirical Taagepera-Nurmia formula describes the alpha half-life as the function of alpha-energy and the atomic number of the daughter nucleus

The theoretical Gamow-Condon-Gurney formula describes the alpha decay constant as a function of the alpha-energy (or rather the $Q$-value of the alpha decay), as well as the nuclear radius and the atomic number of the daughter.

The Gamow-Condon-Gurney formula only applies when the alpha particle is emitted with zero angular momentum. The probability for alpha decay taking away angular momentum is considerably smaller.

5.2. Beta decay

Beta emitters are found all along the stability valley (see Figure 13) starting from $^1_1$H up to $^{253}_{98}$Cf.

If, as a result of $\beta$ decay, excited nuclei are formed, the ground state will be reached through the emission of monoenergetic gamma photons. One might expect, therefore, that $\beta$ radiation, too, is ‘monochromatic’, consisting of electrons with energy $Q_\beta - E_\gamma$ each. In reality, however, $\beta$ spectra are of continuous character with an energy distribution extending from zero to $E_\beta = Q_\beta - E_\gamma$. The energy distribution of the neutrinos is also continuous, being the mirror image of the bell-shaped electron-energy distribution.

This holds both for negative and positive beta decay:

$$\begin{align*}
A \, Z \, X_N \xrightarrow{\beta^-} A \, Z+1 \, X_{N+1}^- + e^- + \bar{\nu}_e, \\
A \, Z \, X_N \xrightarrow{\beta^+} A \, Z-1 \, X_{N+1}^+ + e^+ + \nu_e
\end{align*}$$

(72) (73)

but not for electron capture, the alternative to positive beta decay

$$A \, Z \, X_N \xrightarrow{EC} A \, Z-1 \, X_{N+1}^- + \nu_e$$

(74)

in which monoenergetic neutrinos are emitted since there is no third particle in the final state.

Maximum beta energies ($E_\beta$) span even wider range than alpha energies from the extremely soft 17-18 keV negatron (i.e., $\beta^-$) radiation of $^1_1$H and $^{210}_{82}$Pb, to the very hard 14 MeV positron (i.e., $\beta^+$) radiation of $^8_5$B. The typical $E_\beta$ range—from a few hundred keV to a few MeV—is lower than the usual range of alpha energies (4-7 MeV). Beta half-lives range from fractions of a second for nuclides far from the bottom of the valley of beta stability to billions of years exemplified by $^{40}_{19}$K (1.25×10^9 a) and $^{87}_{37}$Rb (4.7×10^10 a), both of them being important for radioactive dating.

We should mention here the double beta (2$\beta$) decay in which two beta particles and two neutrinos are emitted simultaneously. The typical half-life is on the order of 10^20 a, and therefore such primordial radionuclides can be considered stable for any practical purpose. (Note that the estimated age of our Universe is only about 1.4×10^10 a according to some recent measurements.

The energy dependence of the decay constant for negative $\beta$ decay can be very roughly described by the following empirical formula similar to the Geiger-Nuttal rule:
where \( \alpha > 0 \) and \( \beta \) are constant for separate groups of \( \beta \)-decaying nuclides (Sargent 1933). The Sargent rule, expressed by the above formula, predicts a very steep increase of the decay constant as a function of the beta energy.

The theory of beta decay was developed by Fermi. He found a theoretical relationship describing the dependence of the beta decay constant on beta energy \( E_\beta \) and atomic number \( Z \).

For allowed transitions we can approximately write:

\[
\lambda = \frac{\ln 2}{T_{1/2}} \propto f(Z, E_\beta)
\]

where \( f(Z, E_\beta) \) is the Fermi integral function.

The product

\[
ft \equiv f(Z, E_\beta) T_{1/2}
\]

is called comparative half-life. It is supposed to depend only on the non-trivial nuclear structure effects. It varies in a wide range, depending on the structure of the nucleus and the type of transition.

The Fermi integral function \( f(Z, E_\beta) \) is considered very useful, because it can help visualize the overall dependence of the decay constant \( \lambda \) on the beta energy \( E_\beta \) and the atomic number \( Z \). For instance, we can see from Figure 15 that

1. the probability for beta decay steeply increases with beta energy (this is equally true for positive and negative beta decay),
2. negative beta decay becomes more probable as the atomic number increases,
3. positive beta decay becomes less probable as the atomic number increases.

Friedlander et al. (1981) cite empirical formulae for the energy- and atomic-number dependence of the Fermi integral function. The following formulae (see also Figure 15) are valid in the intervals \( 0 < Z \leq 100 \) and \( 0.1 < E_\beta < 10 \) (where \( Z \) is the atomic number of the daughter and \( E_\beta \) is the maximum of the beta energy):

\[
\log f^- = 4 \log \left( \frac{E_\beta}{\text{MeV}} \right) - 0.005 \log (E_\beta/\text{MeV}) + 0.02 Z + 0.78,
\]

\[
\log f^+ = 4 \log \left( \frac{E_\beta}{\text{MeV}} \right) - 0.009 (Z + 1) \left( \log \frac{E_\beta}{\text{MeV}} \right)^2 - 0.007 Z + 0.79.
\]
FIGURE 15. The Fermi integral function for negative (to the left) and positive beta decay (to the right) calculated from Eqs. (78) and (79). Note that the directions of the Z axes are different in accordance with an opposite trend in the Z dependence of $f$.

FIGURE 16. The Fermi integral function for electron capture calculated from Eq. (80). The figure shows that EC is most likely for larger values of $Z$.

The respective EC formula gives best approximation for $E_\beta > 0.5$ MeV and for low values of $Z$ (see also FIGURE 16):

$$\log f_{EC} = 2 \log (E_\beta / \text{MeV}) + 3.5 \log (Z + 1) - 5.6.$$ (80)

As we have mentioned, electron capture is the alternative process to positive beta decay. For lower decay energies, electron capture often comes out on top from the competition, due to its less demanding energetic condition.

As regards the branching ratio $\lambda_{EC}/\lambda_{\beta^+}$ as a function of $E_\beta$ for different values of the atomic number: for a constant value of $Z$ (i.e., when comparing different isotopes of the same chemical element), larger beta energy favors $\beta^+$ decay. At any given energy, on the other hand,
heavier nuclides prefer to undergo EC. (Note that the larger \( Z \), the larger is the probability to find a shell electron in the nucleus—a necessary condition for electron capture.)

Friedlander et al. (1981) also cite an approximate formula for the branching ratio of L- and K-capture in allowed transitions:

\[
\frac{\lambda_L}{\lambda_K} = (0.0011Z + 0.06) \left( \frac{E_L(\nu)}{E_K(\nu)} \right)^2,
\]

where \( E_L(\nu) \) and \( E_K(\nu) \) are the energy of the neutrino for L- and K-capture, respectively. (The neutrino energy equals \( E_\beta \) less the binding energy of the electron.)

**Gamma decay**

If, for some reason (e.g., as a consequence of \( \alpha \) or \( \beta \) decay), an excited nucleus is formed, the excited state will sooner or later de-excite via gamma decay or alternative processes like internal conversion or the emission of an electron-positron pair. The latter (rather rare) process is called pair emission. (Internal conversion and pair emission are the nuclear analogues of photoelectric effect and pair production.)

In the case of gamma decay,

\[
{}^A_zX + \gamma \rightarrow {}^4X + \gamma,
\]

the excitation energy (or, in other words, the transition energy \( E_T = E_e - E_\gamma \)) is carried away by a gamma photon (\( \gamma \) emission) as well as by the recoil of the nucleus:

\[
E_T = E_\gamma + E_R.
\]

Long-lived excited nuclei \( (T_{1/2} \geq 1\ \mu s) \) are sometimes referred to as excited nuclear isomers and are indicated by a left superscript ‘m’ standing for ‘metastable’:

\[
{}^{Am}_zX \rightarrow {}^4X + \gamma.
\]

In such cases the transition is referred to as isomeric transition (IT).

Excitation may sometimes provide protection against other types of (actual nuclear) decay. We can take as an example the excited isomer \(^{114m}\)In. In this case first de-excitation takes place through gamma emission \( (T_{1/2} = 50\ \text{d}, \ \text{a very long half-life indeed for an excited state}) \), and then the ground-state isomer \(^{114}\)In is transformed by \( \beta^- \) decay \( (T_{1/2} = 72\ \text{s}) \).

It may also occur that, instead of a gamma photon, a shell electron is emitted. This alternative process for nuclear de-excitation is called internal conversion. Electron emission is then followed by the rearrangement of the electronic shell structure, which, in turn, is accompanied by X-ray emission, characteristic of the element concerned, or by the emission of another electron via Auger process.

The kinetic energy of a conversion electron is given by

\[
E_{ce} = E_T - E_b > 0,
\]

where \( E_b \) is the binding energy of the emitted shell electron and \( E_T \) is the transition energy.

Since the binding energy is different for the K-, L-, and M-shells, the kinetic energy of a conversion electron depends on the shell it was emitted from.

The so-called internal conversion coefficient \( (\alpha) \) is defined as the ratio of the probability of electron emission \( (p_e) \) to that of gamma emission \( (p_\gamma) \):
\[ \alpha = \frac{p_e}{p_\gamma} \] (86)

The value of \( \alpha \) is usually between \( 10^{-4} \) and \( 10^2 \). It is calculated as the sum of partial values characteristic of the individual electronic shells:

\[ \alpha = \alpha_K + \alpha_L + \alpha_M + \ldots \] (87)

where:

\[ \alpha_K = \frac{p_e(n=1)}{p_\gamma}; \quad \alpha_L = \frac{p_e(n=2)}{p_\gamma}; \quad \alpha_M = \frac{p_e(n=3)}{p_\gamma}; \ldots \] (88)

and \( n \) is the principal quantum number.

It is an empirical fact that \( \alpha_K > \alpha_L > \alpha_M \ldots \) whenever it is energetically possible to emit K, L, M, ... electrons [see Eq. (85)].

The decay constant (also called by physicists as transition rate) of an excited isomer depends on the energy (\( E_\gamma \)), electric (E) or magnetic (M) character, and on the multipolarity (\( 2^L \); with a common notation: \( E_L \) or \( M_L \)) of the radiation as well as on the initial and final value of the nuclear spin (\( I_i, I_f \)) (and other structural properties of the nucleus).

There are some thumb rules, namely

1. the larger the transition energy, the faster the transition,
2. the lower the multipolarity, the faster the transition (the rule holds for allowed transitions and separately for \( E_L \) and \( M_L \) radiations); if therefore \( E_1 \) (or \( M_1 \)) is allowed, then \( E_2, E_3 \ldots \) (\( M_2, M_3 \ldots \)) can be neglected,
3. for any given multipolarity the electric transition is faster than the magnetic transition,
4. for any given energy, the transition rate decreases in the following order: \( E_1 > M_1 > E_2 > M_2 > E_3 > M_3 > E_4 > M_4 \).

The half-life of excited states (isomers) depends on the change of nuclear spin (\( \Delta I \)) and parity in the transition as well as on the transition energy (\( E_T \)). The greater is \( \Delta I \), the longer is \( T_{1/2} \), but an increase in \( E_T \) causes a decrease in the half-life.

### 5.3. Spontaneous fission

It is known that the fission of heavy nuclei to lighter ones is an energy-producing process above about \( A = 100 \), however actual fission is only observed for \( A \geq 230 \). The reason why the instability of heavy nuclides does not lead to prompt fission automatically lies in the existence of the so-called fission barrier analogous to the potential barrier of alpha decay. Nevertheless, spontaneous fission does exist, in spite of the fission barrier. This can be explained (similarly to \( \alpha \) decay) by tunneling. Since the probability for heavier particles to get across the barrier is less than that for \( \alpha \) particles, the half-life for spontaneous fission is usually much longer than for \( \alpha \) decay. However, some of the heavy transuranium elements fission with relatively short half-lives. \(^{235}\text{U}\) nuclei undergo \( \alpha \) decay with \( T_{1/2} = 7 \times 10^8 \) a and spontaneous fission with \( T_{1/2} = 2 \times 10^{17} \) a, i.e., only one out of 300 million atoms disintegrates by fission. \(^{254}\text{Cf}\), on the other hand, decays only by spontaneous fission with a half-life of just 55 d, which is too short a time to be explained by tunneling.

The partial half-lives for spontaneous fission depend on the fissility parameter \( Z^2/A \). There is a general trend for the half-life to decrease as the fissility parameter increases. However, the lines connecting the points representing the same (even-\( Z \)) element go through a maximum owing to shell effects. Heavier elements (larger \( Z \)) tend to undergo spontaneous
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fission more easily. (As the atomic number increases from $Z = 90$ to $Z = 106$, the partial half-life decreases about 30 orders of magnitude!)

It is interesting to note that when either $Z$ or $N$ is odd, the half-life tends to be much longer. On the other hand, the half-lives of the fission isomers are many orders of magnitude below the general trend.

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