Chapter 1  Radioactivity

The radiations investigated in this course are ionizing radiations. In general, they are classified into

<table>
<thead>
<tr>
<th>Charged particles</th>
<th>Fast electrons</th>
<th>Ions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutral radiations</td>
<td>Photons in ionizing region</td>
<td>Neutrons</td>
</tr>
</tbody>
</table>

1.1. Radioactive decay law

We will start with the concept of radioactivity and the radioactive decay law is the most fundamental principle. What do we mean by a transition (or decay)? A transition occurs when a system changes spontaneously from one state to another.

For our purposes this can be

\[
\text{Radioactive Decay} \quad ^{226}\text{Ra} \rightarrow ^{222}\text{Rn} + \alpha
\]

\[
\text{Atomic de-excitation} \quad \text{Na}^* \rightarrow \text{Na} + \hbar\nu
\]

The decay (or disintegration) constant \( \lambda \) is defined as the decay probability of a nucleus per unit time and its unit is \([\text{s}^{-1}]\) or \([\text{yr}^{-1}]\). One important assumption of radioactive decay is that the decay constant is independent of the age of the nuclei.

For a single isolated system, such as one atom, there is little more to say: if we observe the atom over unit time, \( \lambda \) will be the probability per unit time that it will decay. However, for a group, or an ensemble of atoms, \( \lambda \) becomes a useful quantity.

We have a group of atoms which start off as nuclide \( i \)
They decay to nuclide \( f \) with a decay constant \( \lambda \).

If we start with \( N_0 \) radioactive nuclei \( i \) at \( t = 0 \) and \( N_i(t) \) nuclei are present at time \( t = t \), the number of decay in an time interval \( dt \) is

\[
N_i(t)\lambda dt = -dN_i(t) \quad \Rightarrow \quad N_i(t) = N_0 e^{-\lambda t}
\]

This is the well-known radioactive decay law. The governing equation for the number of nuclei of type \( f \) is

\[
\frac{dN_f(t)}{dt} = \lambda N_i(t)
\]
The half-life $t_{1/2}$ gives the time required for half of nuclei to decay. From its definition, $t_{1/2}$ is related to the decay constant by

$$t_{1/2} = \ln 2 / \lambda = 0.693 / \lambda$$

Another useful life quantity in decay is the mean life (or lifetime) $\tau$, which is defined as the average time that a nucleus can survive before decay. From its definition the mean life becomes

$$\tau = \frac{\int t e^{-\lambda t} dt}{\int e^{-\lambda t} dt} = \frac{1}{\lambda}$$

Therefore, the mean life is simply the inverse of the decay constant. After one mean life, the number of nuclei becomes $1/e$ of the initial number as shown in Fig. 1.1.

From the decay law, we can determine the number of undecayed nuclei at a time $t$. However, measuring $N$ is a very difficult work while measuring the number of decay is much easier since we can detect radiations emitted through decay. If the number of nuclei at time $t_1$ is $N(t_1) = N_0 e^{-\lambda t_1}$ and the number at time $t_2$ is $N(t_2) = N_0 e^{-\lambda t_2}$, the total number of disintegrations in $(t_1, t_2)$ can be expressed as

$$N(t_1) - N(t_2) = N_0 (e^{-\lambda t_1} - e^{-\lambda t_2})$$

When the time interval $\Delta t$ becomes short, the number of disintegrations in $(t, t + \Delta t)$ interval is given by

$$|\Delta N| = N(t) - N(t + \Delta t) = N_0 e^{-\lambda t} (1 - e^{-\lambda \Delta t})$$

If the interval $\Delta t$ is much shorter than the mean life $\tau$, then the $e^{-\lambda \Delta t}$ can be approximated, by the Taylor series expansion, to be

$$e^{-\lambda \Delta t} \approx 1 - \lambda \Delta t$$

and accordingly,

$$|\Delta N| \approx \lambda N_0 e^{-\lambda t} \Delta t$$
Then the differential limit leads to
\[
\frac{dN}{dt} = \lambda N_0 e^{-\lambda t},
\]
which is the definition of the **radioactivity** (or **activity**): the number of disintegrations (or radioactive transitions) per second. If \( \lambda \) is in unit of [s\(^{-1}\)], the activity \( A(t) \) at time \( t \) is
\[
A(t) = \lambda N_0 e^{-\lambda t} = \lambda N(t)
\]
The SI unit of the activity is [Bq], equal to one decay per second. Historically, the curie unit [Ci] has also been used. The relation between two units is
\[
1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}
\]
The specific activity of a radioactive source is defined as its activity per unit mass, [Bq/kg].

### 1.2. Quantum theory of decay

For a nucleus which can undergo decay, let’s assume its nuclear potential is given by \( V + V' \), where \( V \) is the nuclear potential giving the stationary states and \( V' \) is a very weak perturbation potential that can cause transitions between the states. From the perturbation theory in Quantum Mechanics, the general approach is to solve the Schrödinger wave equation for the potential \( V \) (neglecting \( V' \)), which gives static nuclear wave functions. The influence of \( V' \) can be calculated by **Fermi’s Golden Rule**, which gives the transition rate.

For the constant time-independent potential \( V \), let’s assume that solving the Schrödinger equation
\[
-\frac{\hbar^2}{2m} \nabla^2 \psi(\vec{r}) + V(\vec{r})\psi(\vec{r}) = E \psi(\vec{r})
\]
\((\psi(\vec{r}) \text{: Schrödinger wave function, } E \text{: energy})\]
gave the wave function \( \psi_i(\vec{r}) \) of the stationary nuclear states. The complete wave function for the state \( i \) is
\[
\Psi_i(\vec{r}, t) = \psi_i(\vec{r}) e^{-iE_it/\hbar}
\]
where \( E_i \) is the energy of the state \( i \). The corresponding probability of the finding the system in the state \( i \) is \( |\Psi_i(\vec{r}, t)|^2 \), which is independent of time for a stationary state under the potential \( V \) only. The transition probability between the stationary states under the influence of the perturbing potential \( V' \) is calculated by Fermi’s Golden Rule:
\[
\lambda = \frac{2\pi}{\hbar} \left| \int \frac{\hbar}{2} |\psi'_{\beta}(\vec{r})|^2 \right|^2 \rho(E_f) V'_{\beta'} \int \psi^*_{\beta'}(\vec{r}) V' \psi_{\beta} d^3r
\]
where, $\psi_f$ and $E_f$ are the wave function and energy of the final state $f$, $\rho(E_f)$ is the number of accessible final states per unit energy. To be consistent with the decay law, the probability of finding the state $i$ should have $e^{-t/\tau_i}$ dependence on time, which requires the wave function to be modified to:

$$\Psi_i(\vec{r}, t) = \psi_i(\vec{r}) e^{-iE_i t/\hbar} e^{-t/2\tau_i},$$

where $\tau_i = 1/\lambda_i$ is the mean life of the state $i$. Due to the real exponential term, we can no longer determine the energy of the state exactly and the uncertainty of the energy $E_i$ becomes $\Gamma_i = \Delta E_i \approx \hbar / \tau_i$ from the Uncertainty Principle. Therefore, the energy of the state has a distribution given by

$$P(E) = \frac{1}{(E - E_i)^2 + \Gamma_i^2 / 4}$$

Here, $P(E) dE$ means the probability of finding the energy in $(E, E + dE)$ interval in the vicinity of $E_i$. The distribution is shown in Fig. 1.2. The width $\Gamma_i$ represents our inability to determine the energy of the state.

![Energy distribution P(E).](image)

The three primary decay types are $\alpha$, $\beta$, and $\gamma$ decays. In $\alpha$ and $\beta$ decays, an unstable nucleus emits an $\alpha$ or $\beta$ particle so that it becomes a more stable nucleus while an excited state decays to lower energy states in $\gamma$ decays.

<table>
<thead>
<tr>
<th>$\alpha$ decay</th>
<th>$\beta$ decay</th>
<th>$\gamma$ decay</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^A_ZX \rightarrow ^{A-4}_ZX' + ^4He$</td>
<td>$n \rightarrow p + \beta^- + \bar{\nu}$: $\beta^-$ decay</td>
<td>- Competing with the internal conversion</td>
</tr>
<tr>
<td></td>
<td>$p \rightarrow n + \beta^+ + \nu$: $\beta^+$ decay</td>
<td>- $t_{\beta^+} : \sim &lt; 10^{-9}$ s</td>
</tr>
<tr>
<td></td>
<td>$p + e^- \rightarrow n$: electron capture</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>- except for isomeric transition</td>
</tr>
<tr>
<td></td>
<td></td>
<td>($t_{\beta^-}$: hr or day)</td>
</tr>
</tbody>
</table>
1.3. Constant production and decay

The radioactive decay law defined above is for decay alone. However, when radioisotopes are created in research nuclear reactors (neutron irradiation) or accelerators (proton irradiation), both production and radioactive decay are occurring simultaneously. In this case, the rate of the change in number of the radioactive nucleus is given by

$$\frac{dN(t)}{dt} = -\lambda N(t) + K$$

where K is the rate of production and is assumed to be constant. An initial condition $N(t=0) = 0$ gives

$$N(t) = \frac{K}{\lambda} (1 - e^{-\lambda t}) \Rightarrow A(t) = \lambda N(t) = K(1 - e^{-\lambda t})$$

Therefore, if the production rate K is constant, the radioactivity builds up with the irradiation time. If the irradiation time goes to infinity, the radioactivity is eventually saturated to the production rate K, which is also defined as saturated activity. In other word, the production rate is the ultimate limit of radioactivity we can produce.

To calculate the production rate K, we need to begin with a radiation field quantity. An intense radiation field makes the production rate higher. A scalar quantity used to describe radiation field is fluence rate $\phi$ (usually called flux in Nuclear Reactor Theory), which is defined as the number of particles passing a unit area per unit time. It is straightforward that the time integration of the fluence rate is fluence. Dimensions of fluence and fluence rate are [particles/cm$^2$] and [particles/cm$^2$ s], respectively. The fluence rate spectrum $\phi(E)$ represents the energy distribution of the fluence rate and $d\phi = \phi(E)dE$ physically means fluence rate in the energy interval from E to E + dE. In general, a radiation field has a spatial dependence and therefore, the fluence rate spectrum can be represented as $\phi(\vec{r}, E)$.

The production of radioisotopes depends also upon the characteristics of the interaction (or reaction) involved. These are described by the reaction (or interaction) cross-section $\sigma$, which is defined as the reaction probability of a single target nucleus per unit fluence of the radiation field. From its definition, the cross section has a dimension of area and the [barn] unit (1 barn = 10$^{-24}$ cm$^2$) is usually employed. The cross section is strongly dependent on energy of radiation in general.

Combining the radiation field quantity (fluence rate) and the cross section together with the number density of the target nuclide $n_i$ gives the interaction (reaction) rate density:

$$\int \phi(\vec{r}, E)\sigma(E)n_i \, dE$$

The equation means the number of interactions per second per unit volume. The target number density $n_i$ can be obtained from the mass density $\rho$ by
\[ n_t = \theta \frac{\rho}{M} N_A \]

where \( \theta \) is the isotopic abundance of the target nuclide, \( M \) the atomic mass of the target atom, \( \rho \) density of the target atom in [g/cm\(^3\)], and \( N_A \) Avogadro’s number. A convenient unit for \( n_t \) is [atoms/barn-cm] or [\(10^{-24}\) atoms/cm\(^3\)] so that the \(10^{-24}\) term present in the cross section can be canceled out. The interaction rate (production rate here) is obtained by including the volume of the sample:

\[ K = \int \int \phi(r, E) \sigma(E) n_t \, dE \, dV \]

Since we assumed the production rate to be constant, the fluence rate is steady and burn up, decrease in the number of target nuclei, is negligible. If the radiation field is uniform over the sample volume the production rate is simplified to

\[ K = V \int \phi(r, E) \sigma(E) n_t \, dE = \theta \frac{m}{M} N_A \int \phi(E) \sigma(E) \, dE \]

where \( m \) is the mass of the target irradiated. The rate of production \( K \) is proportional to the fluence rate of the radiation field, in which a sample is irradiated.

The rate \( K \) is also proportional to the reaction cross section. The illustration below is just for graphical representation and it does not mean that bigger targets always have larger reaction cross sections.

The rate \( K \) is proportional to the number of target atoms in the sample as well. The more target atoms there are in the sample the greater the number of radioisotopes created, for the same incident particle fluence rate.
Fig. 1.3 shows the activity buildup of $^{28}$Al ($^{27}$Al irradiated with a slow neutron beam) as a function of the irradiation time. The radionuclide $^{28}$Al is produced through $^{27}$Al(n,$\gamma$) reaction in this case. For irradiation times short compared to the half-life of $^{28}$Al, 2.24 min., the exponential term is negligible, which leads to a linear increase in the activity. As the irradiation time becomes longer the growth speed becomes slower. When the irradiation is over, the radioactivity follows the simple decay law. Depending on the irradiation time, the activity at the end of irradiation is smaller or close to the saturated activity.

\[ \begin{array}{c}
\text{Irradiation} \\
\text{Saturated activity} \\
\text{Time [min]} \\
\text{Radioactivity [Bq]} \\
\end{array} \]

Fig. 1.3. $^{28}$Al activity buildup with a constant production rate of $1.2 \times 10^5$ /s.

1.4. Growth of daughter activities

Suppose we have a nuclide of type $A$ that decays to type $B$, and type $B$ decays to type $C$:

\[ A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C \]

Here, $\lambda_A$ and $\lambda_B$ are the decay constants. If $N_A(t)$ and $N_B(t)$ are the number of nuclei of each type present at time $t$, the activities are $\lambda_A N_A(t)$ and $\lambda_B N_B(t)$, respectively. Then the rate of change in the type $B$ becomes
\[
\frac{dN_A(t)}{dt} = \lambda_A N_A(t) - \lambda_B N_B(t)
\]

If the initial condition is \( N_A(t = 0) = N_0 \) and there is no additional supply,

\[
N_A(t) = N_0 e^{-\lambda_A t} \quad \Rightarrow \quad \frac{dN_B(t)}{dt} = \lambda_A N_0 e^{-\lambda_A t} - \lambda_B N_B(t)
\]

From this differential equation we wish to obtain an explicit solution for \( N_B(t) \) as a function of time. The general solution for the initial condition of \( N_B(t = 0) = 0 \) is

\[
N_B(t) = N_0 \frac{\lambda_A}{\lambda_B - \lambda_A} (e^{-\lambda_A t} - e^{-\lambda_B t}) \quad \Rightarrow \quad \lambda_B N_B(t) = \lambda_A N_0(t) \frac{\lambda_B}{\lambda_B - \lambda_A} [1 - e^{-(\lambda_B - \lambda_A) t}]
\]

Depending on the combination of the decay constants, there are several distinct cases.

**A. Daughter longer lived than Parent** \((\lambda_A > \lambda_B)\)

In this case, number of nuclei can be expressed as

\[
N_B(t) = N_0 \frac{\lambda_A}{\lambda_B - \lambda_A} (e^{-\lambda_A t} - e^{-\lambda_B t})
\]

Since the parent decays quickly, the daughter activity rises to a maximum and then decays with its decay constant. If time \( t \) is long enough so that \( e^{-(\lambda_A - \lambda_B) t} \) term be pretty small, the equation becomes

\[
N_B(t) \approx N_0 \frac{\lambda_A}{\lambda_B - \lambda_A} e^{-\lambda_B t}
\]

It is apparent that type B nuclei decay with \( e^{-\lambda_B t} \). Fig. 1.4 shows \( {}^{218}\text{Po} (t_{1/2}=3.1 \text{ min}) \rightarrow {}^{214}\text{Pb} (t_{1/2}=27 \text{ min}) \) decay as an example.

![Fig. 1.4. Decay of \( {}^{218}\text{Po} (t_{1/2}=3.1 \text{ min}) \rightarrow {}^{214}\text{Pb} (t_{1/2}=27 \text{ min}) \).](image)

**B. Daughter shorter lived than parent** \((\lambda_A < \lambda_B)\)

From the solution for the type B, the ratio of the two activities can be expressed as

\[
\frac{\lambda_B N_B(t)}{\lambda_A N_A(t)} = \frac{\lambda_B}{\lambda_B - \lambda_A} [1 - e^{-(\lambda_B - \lambda_A) t}]
\]

In this case, as time \( t \) increases, the exponential term becomes smaller and the activity ratio keeps increasing. Eventually, the activity ratio approaches the limiting value of \( \lambda_B / (\lambda_B - \lambda_A) \), by which the radioactivity of type B becomes bigger than that of type A.

The nuclei of type B decay with the decay constant of type A. This situation is called **transient equilibrium** and is shown in Fig. 1.5 for \( {}^{214}\text{Pb} \rightarrow {}^{214}\text{Bi} \) decay.
C. Daughter much shorter lived than parent \((\lambda_A \ll \lambda_B)\)

If the parent nuclide A is so long-lived and the decay constant \(\lambda_A\) is much bigger than \(\lambda_B\), the activity ratio is further simplified to

\[
\frac{\lambda_B N_B(t)}{\lambda_A N_A(t)} \approx \left[1 - e^{-\lambda_A t}\right]
\]

The time dependence of the activity \(\lambda_B N_B(t)\) is similar to the trend identified in the transient equilibrium, but in this case, the activity \(\lambda_B N_B(t)\) becomes exactly same with the activity \(\lambda_A N_A(t)\) when time \(t\) passed many times of the daughter half-life. In other word, nuclei B are decaying at the same rate at which they are formed. This is called **secular equilibrium**. The time dependence of the activity is very close to that discussed in the section 1.2: “constant production and decay” since the production term of the daughter \(\lambda_A N_A(t)\) is almost constant over the time period of the daughter activity buildup. Fig. 1.6 shows an example of \(^{238}\text{U} \rightarrow ^{234}\text{Th}\) decays.

![Fig. 1.5. Decay of \(^{214}\text{Pb} \rightarrow ^{214}\text{Bi}\).](image1)

![Fig. 1.6. Decay of \(^{238}\text{U} (4.5\text{ billion yr}) \rightarrow ^{234}\text{Th} (24.1\text{ day})\).](image2)

Although there are long-lived natural radioactive nuclides of other varieties, most of those observed today originate with the very heavy elements, which have no stable isotopes at all. These nuclides decay sequentially until a stable nucleus is finally reached. The four series are listed below.

<table>
<thead>
<tr>
<th>Series</th>
<th>Type</th>
<th>Final stable nucleus</th>
<th>Long lived nuclide and (t_1/2) [yr]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thorium</td>
<td>4n</td>
<td>(^{208}\text{Pb})</td>
<td>(^{232}\text{Th}) (1.4\times10^{10})</td>
</tr>
<tr>
<td>Neptunium</td>
<td>4n+1</td>
<td>(^{209}\text{Bi})</td>
<td>(^{237}\text{Np}) (2.1\times10^{6})</td>
</tr>
<tr>
<td>Uranium</td>
<td>4n+2</td>
<td>(^{206}\text{Pb})</td>
<td>(^{238}\text{U}) (4.4\times10^{9})</td>
</tr>
<tr>
<td>Actinium</td>
<td>4n+3</td>
<td>(^{207}\text{Pb})</td>
<td>(^{235}\text{U}) (7.0\times10^{8})</td>
</tr>
</tbody>
</table>
References