Radioactivity

4

Scientific work must not be considered from the point of view of the direct usefulness of it. It must be done for itself, for the beauty of science, and then there is always the chance that a scientific discovery may become like the radium, a benefit.

Marie Curie

4.1 The Discovery

As previously discussed, in 1895 Röntgen discovered x-rays as a result of the fluorescence they caused on a screen coated with barium platinum cyanide noticed that x-rays became fluorescent in the area where the cathode rays (electrons) interacted with the glass.

Two months later, Henry Becquerel, Professor of Physics at the Ecole Polytechnique in Paris, heard about the discovery of x-rays and immediately thought of a possible connection between x-rays and fluorescence. He quickly initiated a series of experiments to test whether fluorescent substances emitted x-rays. In February 1986, he was exploring the possibility that sunlight might cause crystals to emit penetrating rays like the x-rays. As luck would have it, he used uranium–potassium bisulfate (uranium salts were known to be phosphorescent). These crystals were placed next to a photographic plate that was wrapped in dark paper. If sunlight causes the crystals to emit penetrating rays, then these rays might penetrate the paper, and darken the plate. Because the weather was cloudy, Becquerel placed the unwrapped plate in the drawer of a cabinet, but left the crystals on the cabinet. When he developed the plate a couple of days later, to his surprise he found an intense darkening on it, caused by exposure to radiation. He immediately concluded that some invisible rays from the uranium had penetrated the cabinet and the dark paper covering the plate, and finally exposed the photographic plate. For a few years, these rays were known as *Becquerel rays* or *rayons uranique*. Becquerel soon discovered that the rays from the uranium also ionized gases, thus, making them conductors. A crude and simple gold leafed electroscope was used to measure the *activity* of this radiation.

In 1898, Maria Sklodowska Curie and Pierre Curie discovered that thorium also emitted such rays. That year, the phenomenon of penetrating radiation emitted by uranium and thorium was given the name radioactivity by the Curies. Marie observed that the uranium ore, pitchblende, was much more active and discovered that the increased activity was due to the presence of two new radioactive elements, *polonium* and *radium*.

In 1900, the French chemist Paul Villard discovered a form of radioactive emission that was extremely penetrating and was not deflected by a magnetic field analogous to x-rays. This radiation was later given the name, gamma rays.

4.2 Nuclear Disintegration

Ernest Rutherford and his coworkers at Cavendish Laboratory in England distinguished three components in the radiations from natural radioactive elements, which he called α-rays, β-rays, and γ-rays. These components were eventually identified as helium nuclei, electrons and high-energy photons, respectively (Fig. 4.1).

In 1903, Rutherford and Frederick Soddy explained the cause and nature of radioactivity. They discovered that when radioactive atoms emit α or β-rays, they are changed into other varieties of atoms. They also

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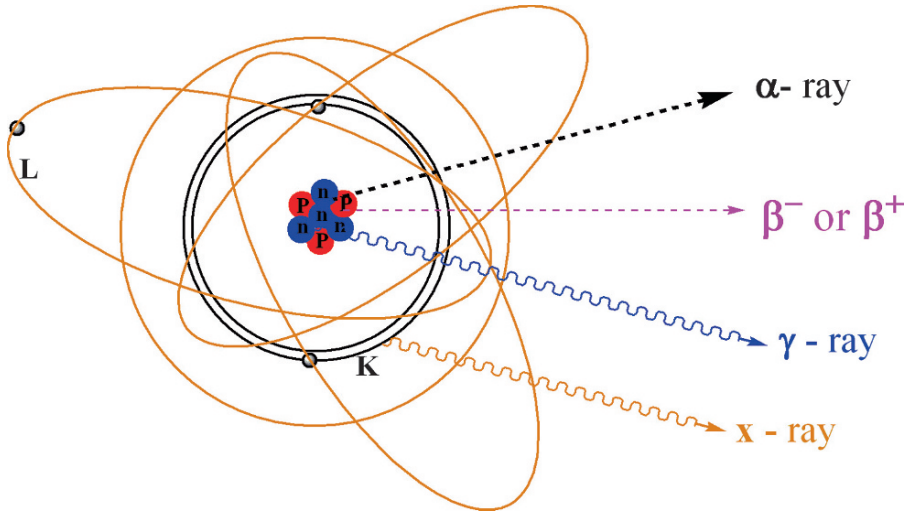

Fig. 4.1 The three components of radioactive emissions originating from the unstable nuclei of natural radionuclides of uranium and thorium: α, β^- and γ, as they were called by Rutherford. Subsequently, two other decay modes, EC and positron emission were identified. In contrast, characteristic x-rays originate from the inner shells of electronic orbits

discovered the existence of atoms of the same element, having different atomic weights. In 1913, Soddy introduced the word *isotopes* to describe atoms of an element that have different atomic weights, but the same chemical properties. They proposed a *nuclear disintegration theory*, which states that radioactivity is actually the changing of one chemical element into another through the emission of α or β particles.

Nuclear transmutation is a phenomenon in which an unstable atomic nucleus disintegrates, in order to acquire a more stable state, by emitting subatomic particles and/or electromagnetic radiation. This phenomenon is called *radioactivity* or *radioactive decay*. One of the most important points to understand is that the energy liberated during the radioactive decay comes from within the individual nuclei of a radioactive element without any external excitation, unlike in the case of atomic radiation.

The emission of *positrons* (β^+) by radioactive isotopes was observed by Irene Curie and Frederick Joliot, in 1934 and they discovered that radioisotopes can be made artificially. With the discovery of nuclear fission, by Otto Hahn in 1939, scientists realized that some unstable nuclides with high atomic numbers may also undergo *spontaneous fission*.

Following the discovery of the proton by Rutherford in 1919, and the neutron by Chadwick in 1932, it became clear that the neutron/proton ratio of any nuclide, and the nuclear binding energy per nucleon (Chap. 3) are the two most important characteristics of any nuclide that would determine the stability of an element, and the kind of radioactive decay it undergoes in order to attain stability.

4.2.1 Types of Radioactive Decay

The radioactive decay processes fall into two categories: one that involves a change in the mass number *A* of a radionuclide, and one in which both the parent and the daughter radionuclides have the same mass number (isobaric decay). The five major types of radioactive decay are shown in Table 4.1. The different radioactive decay modes are shown in the decay scheme diagram (Fig. 4.2).

4.2.1.1 Alpha (α) Decay

Alpha decay (Fig. 4.2) is generally seen in high atomic number elements such as ^{238}U, ^{230}Th and ^{226}Ra. The radionuclide ejects an α particle (two protons and two neutrons) and, because it loses two protons, it is converted into a nuclide with a lower atomic number. Alpha particles are *monoenergetic* and may have kinetic energies in the range of 4–9 MeV. Radionuclides emitting α particles may also emit γ photons. Nuclides with *A*

Table 4.1 Nuclear transmutations

Decaymode	Reason for instability of parent nucleus	Transformation	Example	n/p ratio change
Alpha decay	Too large	${}^{A}_{Z}X \rightarrow {}^{A-4}_{Z-2}Y + {}^{4}_{2}\alpha$	${}^{226}_{88}\mathrm{Ra} \rightarrow {}^{222}_{86}\mathrm{Rn} + {}^{4}_{2}\alpha$	Increases
Beta decay	Neutron rich	${}^{A}_{Z}X \rightarrow {}^{A}_{Z+1}Y + e^{-}$	${}^{14}_{6}\mathrm{C} \rightarrow {}^{14}_{7}\mathrm{N} + e^{-} + v$	Decreases
Positron emission	Neutron deficient	${}^{A}_{Z}X \rightarrow {}^{A}_{Z-1}Y + e^{+}$	${}^{11}_{6}\mathrm{C} \rightarrow {}^{11}_{5}\mathrm{B} + e^{+} + v$	Increases
Electron capture	Neutron deficient	${}^{A}_{Z}X + e^{-} \rightarrow {}^{A}_{Z-1}Y$	${}^{111}_{49}\mathrm{In} + e^{-} \rightarrow {}^{111}_{49}\mathrm{Cd}$	Increases
Isomeric transition	Excess energy	${}^{A}_{Z}X^{*} \rightarrow {}^{A}_{Z}Y + \gamma$	${}^{99m}_{43}\mathrm{Tc}^{*} \rightarrow {}^{99}_{43}\mathrm{Tc} + \gamma$	No change

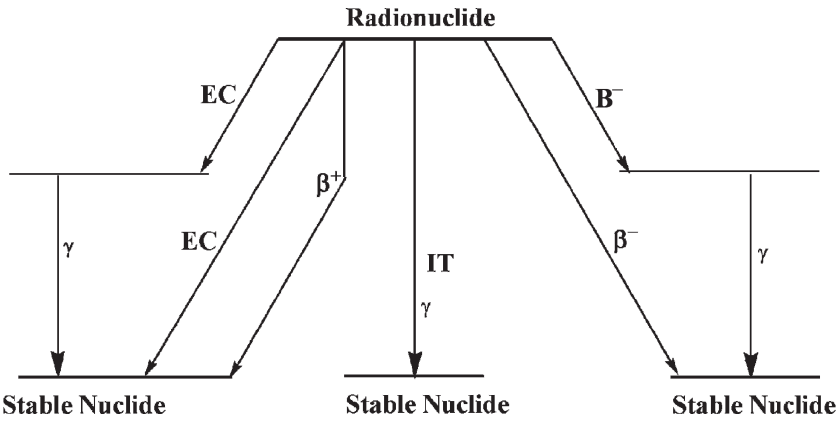

Fig. 4.2 The radioactive decay scheme diagram shows the major decay modes. The parent radionuclide is shown above while the lines to the left represent β^- decay and the lines to the right represent either β^+ decay or EC. Gamma emission is represented by the perpendicular lines. The lines at the bottom represent the ground state energy of the nuclide

>210 are so large, they need to eject α particles in order to reduce their size and become relatively more stable.

$$ {}^{226}_{88}\mathrm{Ra} \rightarrow {}^{222}_{86}\mathrm{Rn} + {}^{4}_{2}\alpha \tag{4.1} $$

4.2.1.2 Beta (β^-) Decay

In beta decay, neutron (*n*)- rich nuclides decay by emitting an electron or β^- rays. In an unstable nucleus, a neutron is converted into a proton (*p*), an electron, and an *antineutrino* (ν). Since only the electron and antineutrino are ejected, the daughter is the next element in the periodic table, with *Z* +1 protons, compared to the parent.

$$ n \rightarrow p + e^{-} + \bar{\nu} \tag{4.2} $$

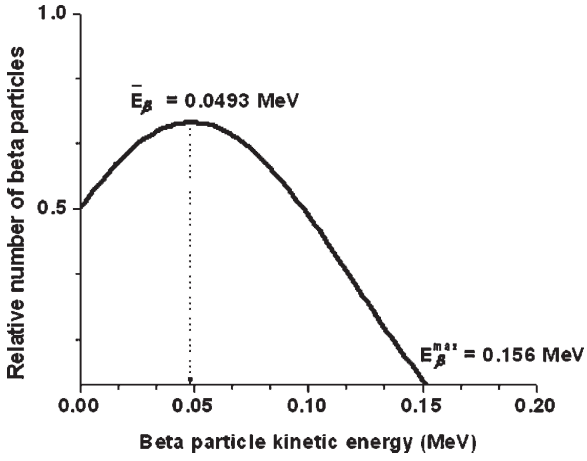

Fig. 4.3 The continuous β^- energy spectrum showing the distribution of β^- particles (relative number versus energy) emitted by ^{14}C. The average energy is denoted by $\bar{E}_\beta$, while the maximum energy is denoted by E_β^{max}

The electron and proton do not exist in the neutron; they together with the neutrino, are created at the moment of decay. The kinetic energies of electrons ejected in the beta decay vary continuously from 0 to a maximum value of KE_{max} (E_{β}^{max}), characteristic of the radionuclide (Fig. 4.3). In an attempt to explain the continuous spectrum of β particle energies, Wolfgang Pauli, in 1930, proposed that a neutral particle with zero or a very small undetectable mass would carry off some of the energy equal to the difference between KE_{max} and the actual kinetic energy of the electron. This hypothetical particle was later called *neutrino* by Enrico Fermi, and was eventually detected in the 1950s by Reines and Cowan. Typically, the average kinetic energy of the β particle ($\bar{E}_{\beta}$), is about one third of E_{β}^{max}. With certain beta-emitting radionuclides, the daughter nuclide in an excited state, which promptly reaches ground state by the emission of γ rays. For example, in case of ^{131}I, 81% of the daughter ^{131}Xe atoms are in an excited state and emit γ photons (364 KeV). Since the emission of γ photons follows almost immediately after the beta emission, this sequential decay process is referred to as a β,γ decay.

4.2.1.3 Positron (β^+) Decay

Nuclides that are deficient in neutrons are unstable, and decrease the number of positive charges in the nucleus (i.e., protons) by decaying either through positron emission or *electron capture* (EC). These two decay modes are regarded as *inverse beta decay*. They are alternatives to reach the ground state when an unstable nucleus is neutron-deficient. Positron emission may be more common in elements with a lower atomic number, while electron capture is seen mostly in elements with high atomic numbers.

A positron is an *antiparticle* of an ordinary electron (Fig. 4.2). In a proton-rich nuclide, a proton in the nucleus is transformed into a neutron, positron, and a neutrino (conversion of a proton into a neutron can only happen inside a nucleus).

$$p \rightarrow n + e^{+} + v \tag{4.3}$$

In 1928, Paul Adrien Maurice Dirac predicted the existence of an antielectron or positron. This prediction was unexpectedly confirmed in 1932 by the American physicist, Carl Anderson, who detected high energy positrons in cosmic radiation.

In 1934, Irene Curie and Frederick Joliot reported the discovery of artificial radioactivity. More specifically, when natural stable ^{27}Al was bombarded with alpha particles, ^{30}P ($T_{½} = 3$ min) was produced, following which positron emission decayed to stable ^{30}Si, as shown below.

$$^{27}_{13}\mathrm{Al} + {}^{4}_{2}\alpha \rightarrow {}^{30}_{15}\mathrm{P} + n \tag{4.4}$$

$$^{30}_{15}\mathrm{P} \rightarrow {}^{30}_{14}\mathrm{Si} + e^{+} + v \tag{4.5}$$

Positron emission leads to a daughter nucleus of a lower atomic number (Z–1), leaving the mass number unchanged. Just as in β^- decay, the positron (β^+) emission spectrum is continuous, (Fig. 4.2) with positron energies ranging from 0 to E_{β}^{max}. The average energy of the positron is 1/3rd of the maximum. The difference between the maximum energy and the actual energy of a positron is carried off by the neutrino. One of the consequences of positron emission is *annihilation radiation*, which is discussed in more detail in Chap. 6. Just as in β^- decay, the daughter nuclide may be in an excited state and as a result, the sequential β^+, γ decay process involves emission of gamma photons from the daughter nucleus. ^{11}C, ^{18}F, ^{64}Cu, ^{68}Ga and ^{124}I, used in developing molecular imaging radiopharmaceuticals for PET, are the most important radionuclides that decay by positron emission.

4.2.1.4 Electron Capture (Ec)

The unstable neutron-deficient nuclides may also reach the ground state by decreasing the positive charge in the nucleus by a process in which an orbital electron, usually from a *K*-shell, is captured by the parent nucleus, resulting in the conversion of one of the protons into a neutron. Since the energy of the parent nuclide is high, a neutrino is also produced, taking away some of the transition energy.

$$p + e^{-} \rightarrow n + v \tag{4.6}$$

The electron capture (*K-capture*) decay process leads to a daughter nucleus of a lower atomic number (Z–1), leaving the mass number unchanged. As a vacancy is created in the *K*-shell of the daughter nuclide, characteristic x-rays and/or Auger electrons are ejected from the daughter nuclide. Sometimes, the daughter nuclide may also be in an excited state due to a change in the *n*/*p* ratio. As a result, gamma photons and conversion

electrons are emitted from the daughter nuclide in a process known as *EC, γ* decay mode. ^{67}Ga, ^{111}In, and ^{123}I are the most important radionuclides with EC decay mode that are useful for developing molecular imaging radiopharmaceuticals for SPECT.

4.2.1.5 Isomeric Transition (IT)

When an unstable decay occurs by any of the decay modes described above, the daughter nucleus may be in an unstable long-lived *metastable* state. Transitions or decays of such metastable nuclides (exist for >10^{1-12} s) result in the emission of gamma photons (Fig. 4.2), but leave both *Z* and *A* unchanged. The two nuclides of an element that differ only in the energy content are called *isomers* (different from the concept of isomers in chemistry). The metastable states are called "isomeric states" (designated with letter *m* next to *A*), and the decay process is known as "isomeric transition." As an alternative, the nucleus of a metastable nucleus may transfer the energy to an orbital electron, also known as a conversion electron, and eject it from the atom. In contrast to beta decay, conversion electrons do not display a continuous energy spectrum, but show discrete energies. The most important radionuclide decaying by isomeric transition is ^{99m}Tc which is very useful for developing molecular imaging agents for use with SPECT.

4.2.1.6 Multiple Decay Mode

Certain radionuclides decay by a single decay mode while some decay by multiple decay modes. As mentioned earlier, neutron deficient radionuclides may decay by EC or by positron emission. However, for a radionuclide to emit both β^- and β^+ particles, the *n/p* ratio must be very close to that of the stable isotope of that element (Table 4.2). Every decay mode may also involve emission of γ photons.

4.2.2 Radioactive Decay Series

Often an unstable radionuclide, especially one with a high atomic number, cannot reach a stable ground state through a single decay process. In such a case a *decay series* occurs until a stable nuclide is formed. A well-known example of this is the decay series that starts with ^{238}U and ends with stable ^{206}Pb. During this process eight alpha particles and six beta particles are emitted before reaching the ground state, after losing ten protons and 22 neutrons.

Table 4.2 Radionuclides and decay modes

					% Decay abundance			
Element	Z	Isotope	n/p	Stability	β^-	β^+	EC	γ
Carbon	6	^{11}C	1.8333	Radioactive	–	99	1	*
		^{12}C	1.0	Stable (98.9%)	–	–	–	–
		^{13}C	1.1666	Stable (1.1%)	–	–	–	–
		^{14}C	1.3333	Radioactive	100	–	–	–
Fluorine	9	^{18}F	1.0	Radioactive	–	97	3	–
		^{19}F	1.1111	Stable (100%)	–	–	–	–
		^{20}F	1.2222	Radioactive	100	–	–	*
Copper	29	^{61}Cu	1.1035	Radioactive	–	100	–	*
		^{62}Cu	1.1379	Radioactive	–	98	2	–
		^{63}Cu	1.1724	Stable (69.17%)	–	–	–	–
		^{64}Cu	1.2069	Radioactive	39	19	41	*
		^{65}Cu	1.2414	Stable (30.83%)	–	–	–	–
		^{67}Cu	1.3103	Radioactive	100	–	–	*
Gallium	31	^{66}Ga	1.1290	Radioactive	–	56	43	*
		^{67}Ga	1.1613	Radioactive	–	–	100	*
		^{68}Ga	1.1935	Radioactive	–	90	10	*
		^{69}Ga	1.2258	Stable (60.1%)	–	–	–	–
		^{71}Ga	1.2903	Stable (39.9%)	–	–	–	–
		^{72}Ga	1.3226	Radioactive	100	–	–	*

With some radionuclides, the series may involve only two or three decays before reaching a stable ground state. For example, the unstable ^{99}Mo reaches stable ^{99}Ru as shown below. The intermediate ^{99}Tc has a long half-life (2.1×10^5 year).

$$^{99}_{42}\mathrm{Mo} \xrightarrow{\beta^-} {}^{99m}_{43}\mathrm{Tc} \xrightarrow{\mathrm{IT}} {}^{99}_{43}\mathrm{Tc} \xrightarrow{\beta^-} {}^{99}_{44}\mathrm{Ru} \qquad (4.7)$$

4.2.3 Nuclear Fission

Nuclear fission is the division or break-up of a heavy unstable nucleus into two lighter nuclei in order to reduce its size. Spontaneous fission may occur in heavy nuclides, but the probability is very low. For example, ^{235}U undergoes spontaneous fission with a very long half-life (2×10^{17} year).

In 1938, Otto Hahn, a German radiochemist, discovered that ^{235}U undergoes fission when struck by a slow neutron. It is not the impact of the neutron that induces fission. Instead, following absorption of the neutron, the ^{236}U nucleus is so unstable that almost at once it explodes into two lighter nuclei known as *fission fragments*. In addition, an enormous amount of energy (≈200 MeV) and 2–3 high energy neutrons, per nuclear fission, are also released. Lise Meitner, a nuclear physicist, coined the name *fission* to describe the splitting of a uranium atom into two lighter elements.

$$^{235}_{92}\mathrm{U} + n \rightarrow {}^{236}_{92}\mathrm{U}^* \xrightarrow{\text{fission}} {}^{140}_{54}\mathrm{Xe} + {}^{94}_{38}\mathrm{Sr} + 2n + \text{Energy} \qquad (4.8)$$

$$^{235}_{92}\mathrm{U} + n \rightarrow {}^{236}_{92}\mathrm{U}^* \xrightarrow{\text{fission}} {}^{141}_{56}\mathrm{Ba} + {}^{92}_{36}\mathrm{Kr} + 3n + \text{Energy} \qquad (4.9)$$

The above processes are only a couple of examples of the many fission fragments that ^{235}U can produce. In fact, over 200 different radio isotopes of 35 different elements have been observed among fission products. Most of these fragments are neutron rich and decay by beta decay.

4.3 Radioactive Decay Equations

4.3.1 Exponential Decay

Radioactive decay or transformation is a statistical process that obeys the laws of chance. No cause–effect relationship is involved in the decay of a radionuclide, only a certain probability per unit time.

The activity (A) of a sample of a radionuclide is the rate at which the nuclei of its constituent atoms (N) decay. Also the activity or the disintegration rate of a radionuclide at any time is proportional to the total number of radioactive atoms present at that time. Mathematically, the rate of decay (A) is given by

$$A = -\frac{dN}{dt} \propto N \qquad (4.10)$$

$$A = -\frac{dN}{dt} \lambda N \qquad (4.11)$$

Where N is the number of radioactive atoms and λ is the decay constant (or proportionality constant) which is defined as the probability of disintegration per unit time. The minus sign indicates that the number of atoms is decreasing with time. The above equation for the rate of decay is the rate law for a first order process. Upon integration, the formula for "radioactive decay" is given by

$$N = N_0 e^{-\lambda t} \qquad (4.12)$$

The formula for the time variation of activity or activity at any time t is given by

$$A_t = A_0 e^{-\lambda t} \qquad (4.13)$$

The above equations represent the *exponential decay* of any radionuclide in which λ, is the *decay constant* of the radionuclide and the factor $e^{-\lambda t}$ is known as the *decay factor*, which is the fraction of original activity A_0 remaining after time t.

The decay factor is an *exponential function* of time t. The exponential decay is characterized by the transformation of a constant fraction of the number of atoms or activity present per unit time interval. When the decay factor, number of atoms, or activity is plotted against time, it is a curve approaching zero on a linear plot, but is a straight line in a semilogarithmic plot (Fig. 4.4).

4.3.2 Units of Activity

The SI unit of radioactivity is named after Becquerel

$$1 \text{ Becquerel} = 1 \text{ Bq} = 1 \text{ decay}/s \qquad (4.14)$$

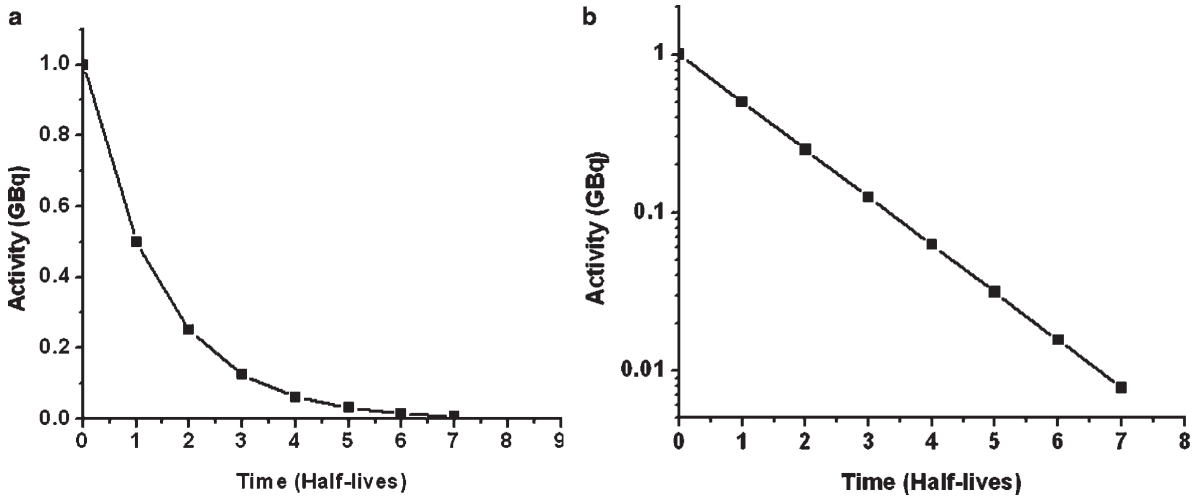

Fig. 4.4 The decay of radioactivity as a function of time (half-lives) plotted on a linear scale (**a**) and semilogarithmic scale (**b**)

The traditional unit of activity is the Curie (Ci), which was originally defined as the activity of one g radium, ^{226}Ra. Curie is now defined as

$$1\ \text{Curie} = 1\ \text{Ci} = 3.7\times10^{10}\ \text{dps} = 37\ \text{GBq} \quad (4.15)$$

$$1\ \text{milliCurie} = 1\ \text{mCi} = 3.7\times10^{7}\ \text{dps} = 37\ \text{MBq} \quad (4.16)$$

$$1\ \text{microCurie} = 1\ \mu\text{Ci} = 3.7\times10^{4}\ \text{dps} = 37\ \text{KBq} \quad (4.17)$$

4.3.3 Half-Life and Average Life Time

Every radionuclide has a characteristic half-life ($T_{½}$), which is the time required for the activity to decay to 50% of its original value. That is, A_0 is reduced to $A_0/2$ in one half-life, $A_0/2^2$ in 2 half-lives, and $A_0/2^n$ in n half-lives. The activity A, of a radionuclide at any time t is given by

$$A_t = \frac{A_0}{2^n} = \frac{A_0}{2^{(t/T_{1/2})}} \quad (4.18)$$

The half-life and the decay constant of a radionuclide are related

$$T_{1/2} = \frac{\ln(0.2)}{\lambda} = \frac{0.693}{\lambda} \quad (4.19)$$

In a given sample of radionuclide, not all atoms decay with the same half-life; some atoms may have a shorter and some atoms may have a longer life span than one half-life. The average, or mean life (τ) of a radionuclide is, therefore, defined as the average of all individual life times of the atoms in a sample of radionuclide experience. The mean life is related to the decay constant and half-life

$$\tau = \frac{1}{\lambda} = \frac{T_{1/2}}{0.693} = 1.44\,T_{1/2} \quad (4.20)$$

4.3.4 Specific Activity

The specific activity (SA) of a radioactive sample is defined as its activity per unit mass. The SA is the ratio of activity to the total mass of the element present. It has units of Ci g^{-1}, Ci mmole^{-1}, or GBq mmole^{-1}.

Many elements have several isotopes and some of the isotopes of an element can also be stable. The element carbon has four isotopes as shown below:

$$^{11}_{6}\text{C},\ ^{12}_{6}\text{C},\ ^{13}_{6}\text{C},\ ^{14}_{6}\text{C}$$

^{12}C and ^{13}C are stable and, in nature, most of the carbon is ^{12}C (99% abundance). ^{14}C ($T_{½}$ = 5,760 year) decays by beta decay while ^{11}C ($T_{½}$ = 20 min) decays by positron emission. When stable isotopes of an element are present along with the radioactive isotope, then the stable isotopes are called *carrier*. When the radioactivity of the sample is said to be with the carrier and the SA of such a radioisotope is low, because the total mass

of the element includes the mass of radioisotope and also that of the carrier. A radioactive sample that does not contain the carrier is called *carrier-free*. The highest possible SA of a radionuclide is its carrier-free SA (CFSA). It is also known as the theoretical SA, and may not necessarily be achieved practically by radionuclide production methods.

CFSA can be calculated since there is a well-defined Activity–Mass relationship based on the following mathematical relationships:

- Based on $T_{½}$, decay constant (λ) can be calculated as $\lambda = 0.693/T_{½}$
- Knowing the activity (Ci or GBq, etc.) and λ, the number of atoms (*N*) in a sample of radioactivity can be determined based on the activity law, $A = \lambda N$
- Based on *N*, the mass (g or moles) of a radioisotope can be determined because one mole (gram atomic weight) of any element contains 6.022×10^{23} atoms (Avogadro's number).

For a number of radioisotopes the carrier-free SA are shown in Table 4.3. The SA calculations for ^{11}C are shown below.

Carrier-free Specific Activity (CFSA) of ^{11}C

1 mole of ^{11}C (11 g) contains 6.022×10^{23} atoms (N)
Since *T*½ of ^{11}C = 20 min,

$$\lambda = \frac{0.693}{20\,\text{Mins} \times 60\text{s}} = 5.775 \times 10^{-5}$$

Activity (Bq) in 1 mole:

$$B_q = 5.775 \times 10^{-5} \times 6.022 \times 10^{23} = 3.478 \times 10^{19}$$

since 1 Ci = 3.7×10^{10} Bq,

$$\frac{3.478 \times 10^{19}}{3.7 \times 10^{10}} = 9.4 \times 10^{8}\,\text{Ci mole}^{-1}$$

since 1 GBq = 10^9 Bq,

$$\frac{3.497 \times 10^{19}}{10^{9}} = 3.478 \times 10^{10}\,\text{GBq mole}^{-1}$$

CFSA of ^{11}C = 9,400 Ci μmole^{-1} or 9.4×10^3 Ci μmole^{-1}

A similar calculation shows that CFSA of $^{14}C = 6.205 \times 10^{-5}$ Ci μmole^{-1}

4.3.5 Serial Radioactive Decay

When a sample of radioactivity consists of a pair of parent and daughter radionuclides, the parent activity (A_p) at any time *t*, is given by the equation 4.13. Because the daughter is continuously produced by the decay of the parent and at the same time the daughter activity is also decaying, estimating the daughter activity (A_d) at any time really depends on the decay constant or the $T_{½}$ of the daughter radionuclide. The *Bateman equa-*

Table 4.3 Radionuclides for PET and SPECT: Half-life and specific activity (SA)[a]

Radionuclides for PET			Radionuclides for SPECT		
Nuclide	$T_{½}$ (min)	SA (Ci μmol^{-1})	Nuclide	$T_{½}$ (min)	SA (Ci μmol^{-1})
^{82}Rb	1.20	150,400			
^{15}O	2.07	91,730			
^{122}I	3.62	51,912			
^{62}Cu	9.76	19,310			
^{13}N	10.0	18,900			
^{11}C	20.4	9,220			
^{94m}Tc	52.0	3,614	^{99m}Tc	360	522
^{68}Ga	68.3	2,766			
^{77}Br	96.0	1,960			
^{18}F	110	1,708			
^{66}Ga	567	331	^{67}Ga	4,320	40
^{64}Cu	768	245			
^{86}Y	884	213	^{111}In	4,020	47
^{89}Zr	4709	39.9			
^{124}I	6,048	31.0	^{123}I	780	237

[a]Maximum theoretical specific activity (SA)

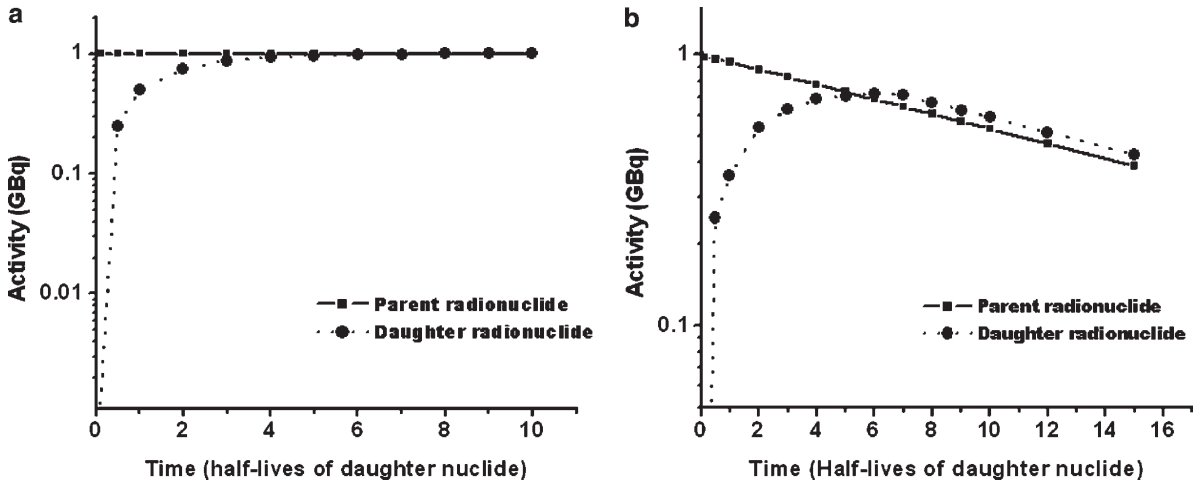

Fig. 4.5 In a parent–daughter decay, following the buildup of the daughter activity, the parent and daughter activities are said to be in secular equilibrium (**a**) or in transient equilibrium (**b**)

tion describes the parent–daughter relationship and provides an estimate of A_d.

$$A_{d(t)} = A_{p(t_0)} \frac{\lambda_d}{\lambda_d - \lambda_p} (e^{-\lambda_p t} - e^{-\lambda_d t}) + A_{d(t_0)} e^{-\lambda_d t} \quad (4.21)$$

At equilibrium, the parent and daughter activities appear to decay with the same rate, even if the two nuclides have different half-lives. Two different equilibrium conditions, however, may exit, depending on how short lived the daughter radionuclide is, compared to the parent. If the parent is short lived compared to the daughter, then there is no equilibrium at all.

4.3.5.1 Secular Equilibrium

When the daughter radionuclide is very short lived, compared to the parent ($T_{½d} << T_{½p}$), *secular equilibrium* is said to exist. The activity of the daughter builds up to that of the parent in about seven half-lives of the daughter. At equilibrium, $A_d = A_p$ (Fig. 4.5 (a)). The activity of the daughter at any time, however, is given by

$$A_{d(t)} = A_{p(t_0)} (1 - e^{-\lambda_d t}) \quad (4.22)$$

Two important examples are ^{68}Ge ($T_{½}$ = 270 days)→^{68}Ga ($T_{½}$ = 68 min) generator and ^{82}Sr ($T_{½}$ = 25 days)→^{82}Rb ($T_{½}$ = 75 s) generator.

4.3.5.2 Transient Equilibrium

When the parent half-life is longer than the daughter half-life ($T_{½p} > T_{½d}$), the daughter activity increases while the parent activity slowly decreases. The daughter activity reaches a maximum and is slightly greater than that of the parent. Once the *transient equilibrium* is reached (~7 $T_{½}$ of daughter), both radionuclides appear to decay with the same rate, the decay rate of the parent (Fig. 4.5(b)). The daughter activity (A_d) at any time t is given by

$$A_{d(t)} = A_{p(t)} \frac{\lambda_d}{\lambda_d - \lambda_p} \quad (4.23)$$

An example of transient equilibrium is ^{99}Mo ($T_{½}$ = 66 h)→^{99m}Tc ($T_{½}$ = 6 h) generator.

Additional Reading

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