CHAPTER

1

ATOMIC STRUCTURE AND RADIOACTIVE DECAY

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OBJECTIVES

By studying this chapter, the reader should be able to:

- Understand the relationship between nuclear instability and radioactive decay.
- Describe the different modes of radioactive decay and the conditions in which they occur.
- Draw and interpret decay schemes.
- Write balanced reactions for radioactive decay.
- State and use the fundamental equations of radioactive decay.
- Perform elementary computations for sample activities.
- Comprehend the principles of transient and secular equilibrium.
- Discuss the principles of the artificial production of radionuclides.

COMPOSITION OF MATTER

The composition of matter has puzzled philosophers for centuries and scientists for decades. Even today the mystery continues, as strange new particles are detected in high-energy accelerators used to probe the structure of matter. Various models proposed to explain the composition and mechanics of matter are useful in certain applications but invariably fall short in others. One of the oldest models, the atomic theory of matter devised by early Greek philosophers,[1] remains today as a useful approach to understanding many physical processes, including those important to the study of the physics of radiation therapy. The atomic model is used in this text, but it is important to remember that it is only a model and that the true composition of matter remains an enigma.

One philosopher, the Reverend George Berkeley (1685–1753), even suggested that matter cannot be proven to exist.

The Atom

The atom is the smallest unit of matter that possesses the physical and chemical properties characteristic of one of the 106 elements, of which 92 occur in nature and the others are produced artificially. The atom consists of a central positive core, termed the *nucleus*, surrounded by a cloud of electrons moving in orbits around the nucleus. The nucleus contains most of the mass of the atom and has a diameter of about 10^{-14} m, whereas the electron cloud, and therefore the atom, has a diameter of about 10^{-10} m. The nucleus contains protons and neutrons. In the neutral atom, the number of protons in the nucleus is balanced by an equal number of electrons in the surrounding orbits. An atom with a greater or lesser number of electrons than the number of protons is termed a *negative* or *positive ion*.

The atomic theory of matter was supplanted over several centuries by the continuum of matter philosophy of Aristotle and the Stoic philosophers. The atomic theory was revived in 1802 when Dalton developed the Principle of Multiple Proportions.

Protons are particles with a mass of 1.6734×10^{-27} kg and a positive charge of $+1.6 \times 10^{-19}$ coulomb.

Neutrons are particles with a mass of 1.6747×10^{-27} kg and no electrical charge.

Electrons have a mass of 9.108×10^{-31} kg and a negative charge of -1.6×10^{-19} coulomb.

In 1999, physicists at the Lawrence Berkeley Laboratory announced the creation of a new element with 118 protons ($Z = 118$), which then decayed to other new elements ($Z = 116$, $Z = 114$, $Z = 112$, etc.) down to the element with 106 protons ($Z = 106$). In 2001, this announcement was retracted.

An atom can be characterized by the symbolism ${}^{A}_{Z}X$, where A is the number of nucleons (i.e., the number of protons plus the number of neutrons) in the nucleus, Z is the number of protons in the nucleus (or the number of electrons in the neutral atom), and X represents the chemical symbol for the particular element to which the atom belongs. The number of nucleons A is termed the *mass number* of the atom, and Z is called the *atomic number* of the atom. The difference $A - Z$ is the number of neutrons in the nucleus, termed the *neutron number* N. Each element has a characteristic atomic number but can have several mass numbers depending on the number of neutrons N in the nucleus. For example, the element hydrogen has the unique atomic number of 1, signifying the solitary proton that constitutes the hydrogen nucleus, but can have none $\left({}^{1}_{1}H\right)$, one $\left({}^{2}_{1}H\right)$, or two $\left({}^{3}_{1}H\right)$ neutrons. The atomic forms ^{1}H, ^{2}H, and ^{3}H (the subscript 1 can be omitted because it is redundant with the chemical symbol) are said to be isotopes of hydrogen because they contain different numbers of neutrons combined with the single proton characteristic of hydrogen. Isotopes of an element have the same Z but different values of A, reflecting a different neutron number N. Isotones have the same N but different values of Z and A. ^{3}H, ^{4}He, and ^{5}Li are isotones because each nucleus contains two neutrons ($N = 2$). Isobars have the same A but

different values of Z and N. ^{3}H and ^{3}He are isobars ($A = 3$). Isomers are different energy states of the same atom and therefore have identical values of Z, N, and A. For example ^{99m}Tc and ^{99}Tc are isomers because they are two distinct energy states of the same atom. The m in ^{99m}Tc signifies a metastable energy state that exists for a finite time (6 hours half-life) before changing to ^{99}Tc. The term *nuclide* refers to an atom in any form.

ISOTOPES have the same number of PROTONS.
ISOTONES have the same number of NEUTRONS.
ISOBARS have the same mass number *A*.
ISOMERS are different ENERGY states of the same atom.

Atomic Units

Units employed to describe dimensions in the macroscopic world are too large to use at the atomic level. Units more appropriate for atomic processes include the atomic mass unit (amu) for mass, electron volt (eV) for energy, nanometer (nm) for distance, and electron charge (e) for electrical charge.

The *amu* is defined as $^{1}/_{12}$ of the mass of an atom of the most common form of carbon, ^{12}C, which has 6 protons, 6 neutrons, and 6 electrons. By definition, the atomic mass of an atom of ^{12}C is 12.00000 amu. In units of amu, the masses of atomic particles are:

$$\begin{aligned} \text{electron} &= 0.00055 \text{ amu} \\ \text{proton} &= 1.00727 \text{ amu} \\ \text{neutron} &= 1.00866 \text{ amu} \end{aligned}$$

One amu $= 1.66 \times 10^{-27}$ kg.

Every atom has a characteristic atomic mass A_m. The gram-atomic mass of an isotope is an amount of the isotope in grams that is numerically equivalent to the isotope's atomic mass. For example, one gram-atomic mass of ^{12}C is exactly 12 grams. One gram-atomic mass contains 6.0228×10^{23} atoms, a constant value that is known as Avogadro's number N_A. With these expressions, the following variables can be computed:

$$\begin{aligned} \text{Number atoms/g} &= N_A/A_m \\ \text{Number electrons/g} &= (N_A Z)/A_m \\ \text{Number g/atom} &= A_m/N_A \end{aligned}$$

The electron volt (eV) is a unit of energy equal to the kinetic energy of a single electron accelerated through a potential difference (voltage) of 1 volt. One keV $= 10^3$ eV, and 1 MeV $= 10^6$ eV. One nanometer (nm) is 10^{-9} meters. The electron unit of electrical charge $= 1.6 \times 10^{-19}$ coulombs.

Count Amadeo Avogadro (1776–1856) was an Italian physicist and chemist.

One eV is equal to 1.6×10^{-19} joule of energy.

The angstrom (Å) unit of atomic distance, equal to 10^{-10} m and widely employed in the past, is seldom used today.

Example 1-1

What is the kinetic energy (E_k) of an electron accelerated through a potential difference of 400,000 volts (400 kilovolts [kV])?

$$\begin{aligned} E_k &= (1 \text{ electron})(400{,}000 \text{ volts}) \\ &= 400{,}000\,\text{eV} = 400\,\text{keV} \end{aligned}$$

Mass Defect and Binding Energy

The neutral ^{12}C atom contains 6 protons, 6 neutrons, and 6 electrons. The mass of the components of this atom can be computed as:

$$\begin{aligned} \text{Mass of 6 protons} &= 6(1.00727 \text{ amu}) = 6.04362 \text{ amu} \\ \text{Mass of 6 neutrons} &= 6(1.00866 \text{ amu}) = 6.05196 \text{ amu} \\ \text{Mass of 6 electrons} &= 6(0.00055 \text{ amu}) = 0.00330 \text{ amu} \\ \hline \text{Mass of components of } ^{12}\text{C} &= 12.09888 \text{ amu} \end{aligned}$$

The equivalence of mass and energy [$E = mc^2$] is arguably the most notable of Einstein's many contributions to science.

The mass of an atom of ^{12}C, however, is 12.00000 amu by definition. That is, the sum of the masses of the components of the ^{12}C atom exceeds the actual mass of the atom, and there is a *mass defect* of 0.09888 amu in the ^{12}C atom. This mass must be supplied to separate the ^{12}C atom into its constituents. The mass defect can be satisfied by supplying energy to the atom according to Einstein's expression $E = mc^2$ for the equivalence of mass and energy. In this expression, E is energy, m is mass, and c is the constant speed of light in a vacuum (3×10^8 m/sec). From the formula for mass-energy equivalence, 1 amu of mass is equivalent to 931 MeV of energy. For example, the energy equivalent to the mass of the electron is (0.00055 amu) (931 MeV/amu) = 0.51 MeV.

The energy associated with the mass defect of ^{12}C is (0.09888 amu)(931 MeV/amu) = 92.0 MeV. The energy equivalent to the mass defect of an atom is known as *the binding energy of the atom* and is the energy required to separate the atom into its constituent parts. Almost all of the binding energy of an atom is associated with the nucleus and reflects the influence of the strong nuclear force that binds particles together in the nucleus. For ^{12}C, the average binding energy per nucleon is 92.0 MeV/12 = 7.67 MeV/nucleon.

Four forces are thought to exist in nature. In order of increasing strength, they are the (1) gravitational force, (2) electrostatic force, (3) weak force, and (4) nuclear force. An explanation of the common origin of these forces is the objective of the Grand Unified Theory of theoretical physics.

In computing the average binding energy per nucleon as the quotient of the binding energy of the atom divided by the number of nucleons, the small contribution of electrons to the binding energy of the atom is ignored.

Example 1-2

What is the average binding energy per nucleon of ^{16}O with an atomic mass of 15.99492 amu?

$$\begin{aligned}
\text{Mass of 8 protons} &= 8(1.00727 \text{ amu}) = 8.05816 \text{ amu} \\
\text{Mass of 8 neutrons} &= 8(1.00866 \text{ amu}) = 8.06928 \text{ amu} \\
\text{Mass of 8 electrons} &= 8(0.00055 \text{ amu}) = 0.00440 \text{ amu} \\
\hline
\text{Mass of components of } ^{16}O &= 16.13184 \text{ amu} \\
\text{Mass of } ^{16}O \text{ atom} &= 15.99492 \text{ amu} \\
\text{Mass defect} &= 16.13184 \text{ amu} - 15.99492 \text{ amu} \\
&= 0.13692 \text{ amu} \\
\text{Binding energy of } ^{16}O &= (0.13692 \text{ amu})(931 \text{ MeV/amu}) \\
&= 127.5 \text{ MeV} \\
\text{Average binding energy per nucleon} &= (127.5 \text{ MeV})/16 \\
&= 7.97 \text{ MeV/nucleon}
\end{aligned}$$

Uncontrolled nuclear fission is the process employed in a uranium or plutonium atomic ("A") bomb. Controlled nuclear fission is the process employed in a nuclear reactor. Uncontrolled nuclear fusion is the process employed in a fusion ("hydrogen") bomb. Efforts to develop controlled fusion have not succeeded so far.

The average binding energy per nucleon is plotted in Figure 1-1 as a function of the mass number of different isotopes. The greatest average binding energies per nucleon occur for isotopes with mass number in the range of 50 to 100. Heavier isotopes gain binding energy by splitting into lighter isotopes. This is equivalent to saying that heavier isotopes release energy when they split into lighter isotopes, a process known as *nuclear fission*. The isotopes ^{233}U, ^{235}U, and ^{239}Pu fission spontaneously when a neutron is added to the nucleus. This process is the origin of the energy released during fission in nuclear reactors and fission weapons. Similarly, energy is released when light isotopes combine to form products with higher average binding energies per nucleon. This latter process is termed *nuclear fusion* and is the source of energy released during a fusion reaction such as that in a "hydrogen" bomb. Controlled nuclear fusion that permits its use for constructive purposes has so far eluded research efforts.

Electron Energy Levels

The model of the atom in which electrons revolve in orbits around the nucleus was developed by Niels Bohr in 1913.[2] This model represented a departure from explanations of the atom that relied on classical physics. In the Bohr model, each orbit or "shell" can hold a maximum number of electrons defined as $2n^2$, where n is the number of the electron shell. The first ($n = 1$ or K) shell can hold up to 2

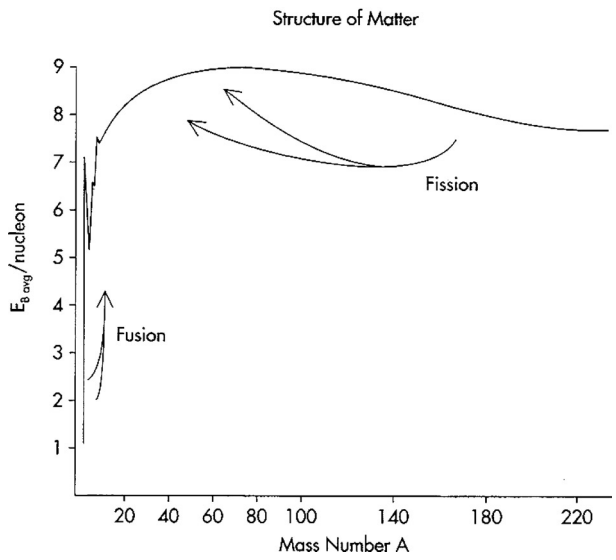


FIGURE 1-1
Average binding energy per nucleon versus mass number.

electrons, the second ($n = 2$ or L) shell can contain up to 8 electrons, the third ($n = 3$ or M) shell can hold up to 18 electrons, and so on. The outermost occupied M, N, or O electron shell, however, can hold no more than 8 electrons, and additional electrons begin to fill the next level to create a new outermost shell before more than 8 electrons are added to an M or higher shell. The number of valence electrons in the outermost shell determines the chemical properties of the atom and the elemental species to which it belongs. Examples of electron orbits in representative atoms are shown in Figure 1-2.

An electron neither gains nor loses energy so long as it remains in a specific electron orbit. Energy is needed, however, to move an electron from one orbit to another farther from the nucleus because work must be done against the attractive electrostatic force of the positive nucleus for the negative electron. Similarly, energy is released when an electron moves from one orbit to another nearer the nucleus. This transition can occur only if a vacancy exists in the nearer orbit, perhaps because an electron has been ejected from that orbit by some physical process. The energy required to remove an electron completely from an atom is defined as the *binding energy of the electron*. The positive charge of the nucleus (i.e., the Z of the atom) and the particular shell from which the electron is removed are the principal influences on the electron's binding energy. Minor influences are the particular energy subshell of the electron within the orbit and the direction of rotation as the electron spins on

"Bohr's work on the atom was the highest form of musicality in the sphere of thought."

A. Einstein as quoted in R. Moore: *Niels Bohr. The Man, His Science and the World They Changed.* Alfred Knopf, New York, 1966.

The maximum number of electrons in a particular electron orbit is defined by the Pauli Exclusion Principle, which states that in any atom (or atomic system), no two electrons can have the same four quantum numbers. The four quantum numbers of an electron are the *principal, azimuthal, magnetic*, and *spin* quantum numbers.

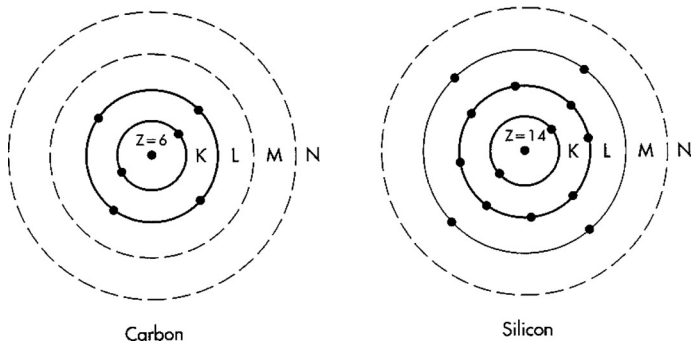
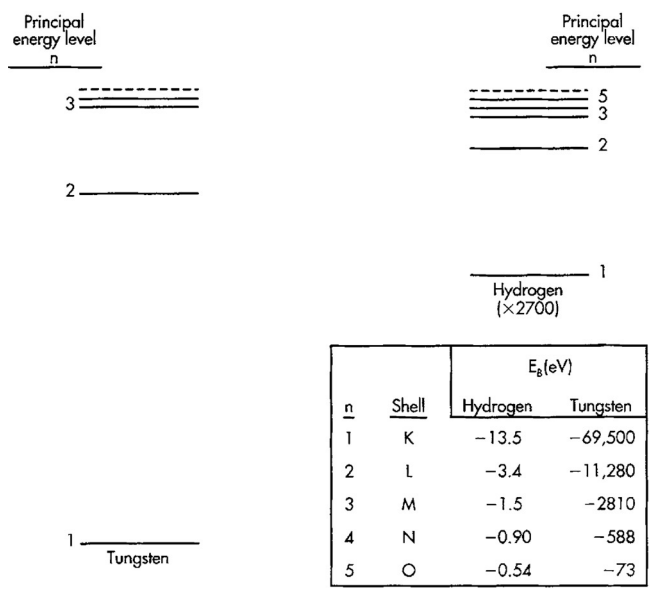


FIGURE 1-2
Electron "orbits" in the Bohr model of the atom for carbon ($Z = 6$) and silicon ($Z = 14$).



		E_B(eV)	
n	Shell	Hydrogen	Tungsten
1	K	−13.5	−69,500
2	L	−3.4	−11,280
3	M	−1.5	−2810
4	N	−0.90	−588
5	O	−0.54	−73

FIGURE 1-3
Binding energies for electrons in hydrogen ($Z = 1$) and tungsten ($Z = 74$). A change in scale is required to show both energy ranges in the same diagram.

its own axis while it revolves in the electron orbit. The electron orbits of a particular atom can be characterized in terms of the binding energies of electrons in the orbits.

Binding energies for electron orbits in hydrogen ($Z = 1$) and tungsten ($Z = 74$) are compared in Figure 1-3. Binding energies are much greater in tungsten than in hydrogen because the higher nuclear charge exerts a stronger attractive force on the electrons. In hydrogen, an electron moving to the K shell from a level farther from the nucleus releases energy usually in the form of ultraviolet radiation. In tungsten, an electron falling into the K shell releases energy usually in the form of an x ray, a form of electromagnetic radiation much more energetic than ultraviolet radiation. The actual energy released equals the difference in binding energy between the electron orbits representing the origin and destination of the electron. For example, an electron moving from the L to the K shell in tungsten releases (69,500 – 11,280 = 58,220 eV = 58.2 keV) of energy, whereas an electron falling from the M to the K shell in tungsten releases (69,500 – 2810 = 66,690 eV = 66.7 keV). X rays emitted by electron transitions between orbits are termed characteristic x rays because their energy is defined by the atomic number of the atom and the particular electron shells involved in the transition.

Characteristic x rays are sometimes called “fluorescence x rays.”

X rays from electron transitions from the L to the K shell are termed K_α x rays. X rays resulting from M to K transitions are termed K_β x rays. Similarly, L_α x rays result from M to L transitions, and L_β results from N to L transitions, and so on.

The physicist H. G. J. Moseley studied the x-ray spectra from 38 elements and used his results to refine the Bohr model of the atom. Moseley was killed in 1915 in the ill-fated Dardanelles expedition of World War I.

When an electron falls from the L to the K shell in a heavy atom, a vacancy is created in the L shell. This vacancy is usually filled instantly by an electron from a shell farther from the nucleus, usually the M shell. The vacancy created in this shell is then filled by another electron from a more distant orbit. Hence, a vacancy in an inner shell of an atom usually results in a cascade of electrons with the emission of a range of characteristic energies, often as electromagnetic radiation. In tungsten, transitions of electrons into the K and L shell result in the release of x rays, whereas transitions into *M* and higher shells produce radiations too low in energy to qualify as x rays.

Energy liberated as an electron falls to an orbit closer to the nucleus is not always released as electromagnetic radiation. Instead, it may be transferred to another electron farther from the nucleus, resulting in the ejection of the electron from its orbit. The ejected electron is termed an *Auger electron* and has a kinetic energy equal to the energy transferred to it, decreased by the binding energy required to eject the electron from its orbit. For example, an electron falling from the L to the K shell in tungsten releases 58,220 eV of energy. If this energy is transferred to another electron in the L shell this electron is ejected with a kinetic energy of (58,220 – 11,280 =

46,940) eV. Usually an Auger electron is ejected from the same energy level that gave rise to the original transitioning electron. In this case, the kinetic energy of the Auger electron is $E_{bi} - 2E_{bo}$, where E_{bi} is the binding energy of the inner electron orbit that receives the transitioning electron, and E_{bo} is the energy of the orbit that serves as the origin of both the transitioning and the Auger electrons.

Example 1-3

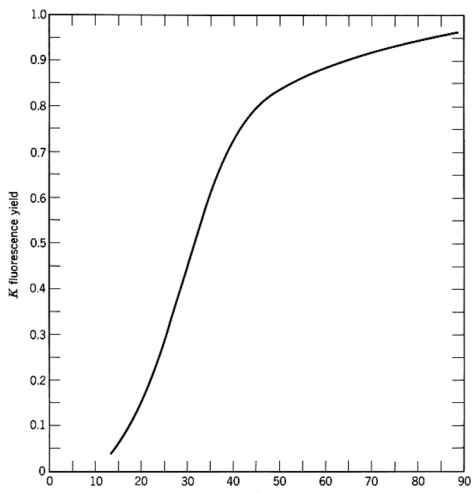
What is the kinetic energy E_k of an Auger electron released from the L shell of gold [$(E_b)_L = 13.335$ keV] as an electron falls from the L to the K shell [$(E_b)_K = 80.713$ keV] in gold?

$$E_k = E_{bi} - 2E_{bo} = [80.713 - 2(13.335)]\text{keV}$$
$$= 54.043\,\text{keV}$$

The emission of characteristic electromagnetic radiation and the release of Auger electrons are alternative processes that release energy from an atom during electron transitions. The *fluorescence yield w* defines the probability that an electron vacancy will result in the emission of characteristic radiation as it is filled by an electron from a higher orbit.

$$w = \frac{\text{Number of characteristic radiations emitted}}{\text{Number of electron shell vacancies}}$$

For low-Z nuclides, Auger electrons tend to be emitted more frequently than characteristic radiations, as shown in Margin Figure 1-1. As Z increases, the fluorescence yield also increases, so that characteristic radiations are released more frequently than Auger electrons.[3]



MARGIN FIGURE 1-1
K-shell fluorescence yields as a function of atomic number.[14]

Nuclear Stability

The nuclei of many atoms are stable. In general, it is these atoms that constitute ordinary matter. In stable nuclei of lighter atoms, the number of neutrons is about equal to the number of protons. A high level of symmetry exists in the placement of protons and neutrons into nuclear energy levels similar to the electron shells constituting the extranuclear structure of the atom. The assignment of nucleons to energy levels in the nucleus is referred to as the "shell model" of the nucleus. For heavier stable atoms, the number of neutrons increases faster than the number of protons, suggesting that the higher energy levels are spaced more closely for neutrons than for protons. The number of neutrons (i.e., the neutron number) in nuclei of stable atoms is plotted in Figure 1-4 as a function of the number of protons (i.e., the atomic number). Above $Z = 83$, no stable forms of the elements exist, and the plot depicts the neutron/proton (N/Z) ratio for the least unstable forms of the elements (i.e., isotopes that exist for relatively long periods before changing).

Nuclei that have an imbalance in the N/Z ratio are positioned away from the stability curve depicted in Figure 1-4. These unstable nuclei tend to undergo changes within the nucleus to achieve more stable configurations of neutrons and protons. The changes are accompanied by the emission of particles and electromagnetic radiation (photons) from the nucleus, together with the release of substantial amounts of energy related to an increase in binding energy of the nucleons in their final nuclear configuration. These changes are referred to as *radioactive decay* of the nucleus, and the process is described as *radioactivity*. If the number of protons is different between the initial and final nuclear configurations, Z is changed and the nucleus is transmuted from one elemental form to another. The various processes of radioactive decay are summarized in Table 1-1.

Additional models of the nucleus have been proposed to explain other nuclear properties. For example, the "liquid drop" (also known as the "collective") model was proposed by the Danish physicist Niels Bohr[3] to explain nuclear fission. The model uses the analogy of the nucleus as a drop of liquid.

Radioactivity was discovered in 1896 by Henri Becquerel[4] who observed the emission of radiation (later shown to be beta particles) from uranium salts. A sentence from his 1896 publication reads "We may then conclude from these experiments that the phosphorescent substance in question emits radiations which penetrate paper opaque to light and reduces the salts of silver." Becquerel experienced a skin burn from carrying a radioactive sample in his vest pocket. This is the first known bioeffect of radiation exposure.

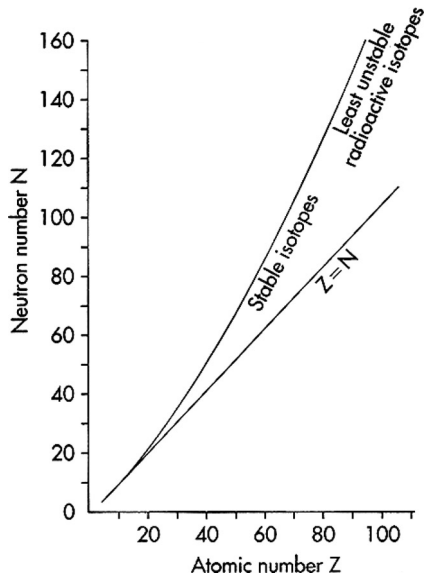


FIGURE 1-4
Number of neutrons (N) in stable (or least unstable) nuclei as a function of the number of protons (atomic number Z).

RADIOACTIVE DECAY

Radioactivity can be described mathematically without reference to the specific mode of decay of a sample of radioactive atoms. The rate of decay (the number of atoms decaying per unit time) is directly proportional to the number of radioactive atoms N present in the sample:

$$\Delta N/\Delta t = -\lambda N \qquad \textbf{(1-1)}$$

Equation (1-1) describes the expected decay rate of a radioactive sample. At any moment the actual decay rate may differ somewhat from the expected rate because of statistical fluctuations in the decay rate.

Equation (1-1) depicts a reaction known as a first-order reaction.

The decay constant λ is also called the *disintegration constant*.

where $\Delta N/\Delta t$ is the rate of decay. The constant λ is the *decay constant* of the particular species of atoms in the sample, and the negative sign reveals that the number of radioactive atoms in the sample is diminishing as the sample decays. The decay constant can be expressed as $-(\Delta N/\Delta t)/N$, revealing that it represents the fractional rate of decay of the atoms. The value of λ is characteristic of the type of atoms in the sample and changes from one nuclide to the next. Units of λ are $(\text{time})^{-1}$. Larger values of λ characterize more unstable nuclides that decay more rapidly.

The decay constant of a nuclide is truly a constant; it is not affected by external influences such as temperature and pressure, or by magnetic, electrical, or gravitational fields.

The rate of decay of a sample of atoms is termed the *activity A* of the sample (i.e., $A = \Delta N/\Delta t$). A rate of decay of 1 atom per second is termed an *activity* of *1 becquerel (Bq)*. That is,

$$1\ \text{Bq} = 1\ \text{disintegration per second (dps)}$$

The "deltas" in $\Delta N/\Delta t$ signify a very small change in the number of atoms occurring over a very small increment of time Δt. The rate of decay is often written in differential form as dN/dt.

The rutherford (Rf) was once proposed as a unit of activity, where $1\ \text{Rf} = 10^6$ dps. The Rf did not gain acceptance in the scientific community and eventually was abandoned. In so doing, science lost an opportunity to honor one of its pioneers.

TABLE 1-1 Radioactive Decay Processes

Type of Decay	*A′*	*Z′*	*N′*	*Comments*
Negatron ($\beta-$)	A	$Z+1$	$N-1$	$E_{\beta\text{-mean}} \cong \frac{E_{max}}{3}$
Positron (β^+)	A	$Z-1$	$N+1$	$E_{\beta\text{-mean}} \cong \frac{E_{max}}{3}$
Electron capture	A	$Z-1$	$N+1$	Characteristic + auger electrons
Isomeric transition gamma (γ) emission	A	Z	N	Metastable if $T_{1/2} > 10^{-6}$ sec
Internal conversion (IC)	A	Z	N	IC electrons: characteristic + auger electrons
Alpha (α)	$A-4$	$Z-2$	$N-2$	

A common unit of activity is the megabecquerel (MBq), where 1 MBq $= 10^6$ dps. An earlier unit of activity, the curie (Ci) is defined as

$$1\,\text{Ci} = 3.7 \times 10^{10}\ \text{dps}$$

The curie was defined in 1910 as the activity of 1 g of radium. Although subsequent measures revealed that 1 g of radium has a decay rate of 3.61×10^{10} dps, the definition of the curie was left as 3.7×10^{10} dps.

Multiples of the curie are the picocurie (10^{-12} Ci), nanocurie (10^{-9} Ci), microcurie (10^{-6} Ci), millicurie (10^{-3} Ci), kilocurie (10^3 Ci), and megacurie (10^6 Ci). The becquerel and the curie are related by 1 Bq = 1 dps $= 2.7 \times 10^{-11}$ Ci. The activity of a radioactive sample per unit mass (e.g., MBq/mg) is known as the *specific activity* of the sample.

Example 1-4

A. $^{60}_{27}$Co has a decay constant of 0.131 y^{-1}. Find the activity in MBq of a sample containing 10^{15} atoms.

$$A = \lambda N = \frac{(0.131\ y^{-1})(10^{15}\ \text{atoms})}{31.54 \times 10^6\ \text{sec/y}}$$
$$= 4.2 \times 10^6\ \text{atoms/s} = 4.2 \times 10^6\ \text{Bq}$$
$$= 4.2\ \text{MBq}$$

B. What is the specific activity of the sample in MBq/g? The gram-atomic mass of ^{60}Co is 59.9338.

$$\text{Sample mass} = \frac{(10^{15}\ \text{atoms})(59.9338\ \text{g/g-atomic mass})}{6.023 \times 10^{23}\ \text{atoms/g-atomic mass}}$$
$$= 9.95 \times 10^{-8}\ \text{g}$$
$$\text{Specific activity} = (4.2\ \text{MBq})/(9.95 \times 10^{-8}\ \text{g})$$
$$= 42 \times 10^6\ \text{MBq/g}$$

Through the process of mathematical integration, an expression for the number N of radioactive atoms remaining in a sample after a time t has elapsed can be shown to equal:

$$N = N_0 e^{-\lambda t} \qquad \textbf{(1-2)}$$

Equation (1-2) reveals that the number N of parent atoms decreases *exponentially* with time.

where N_0 is the number of atoms present at time $t = 0$. This expression can also be written as:

$$A = A_0 e^{-\lambda t} \qquad \textbf{(1-3)}$$

where A is the activity of the sample at time t, and A_0 is the activity at time $t = 0$.

The number of radioactive atoms N^* that have decayed after time t is $N_0 - N$ or

$$N^* = N_0(1 - e^{-\lambda t}) \qquad \textbf{(1-4)}$$

The probability that a particular atom will not decay during time t is N/N_0 or $e^{-\lambda t}$, and the probability that the atom will decay during time t is $1 - N/N_0$ or $1 - e^{-\lambda t}$. For small values of λt, the probability of decay $(1 - e^{-\lambda t})$ can be approximated as λt or, expressed as the probability of decay per unit time, p(decay per unit time) $\sim \lambda$.

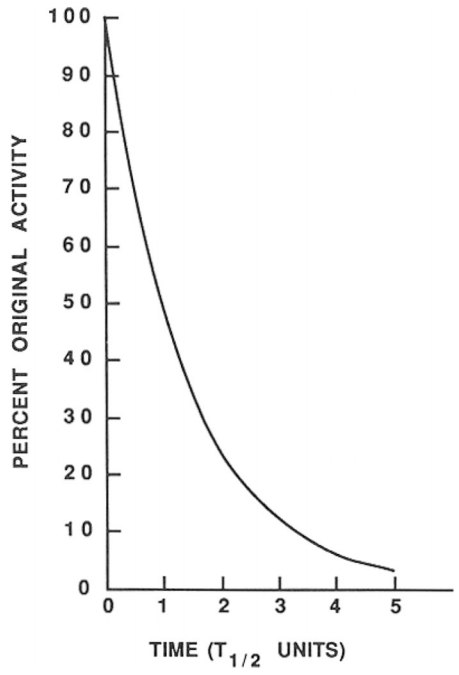
Radioactive decay must always be described in terms of the probability of decay; whether any particular radioactive nucleus will decay within a specific time period is never certain.

The *physical half-life* $T_{1/2}$ of a radioactive sample is the time required for half of the atoms in the sample to decay. The half-life is related to the decay constant of the sample through the expression

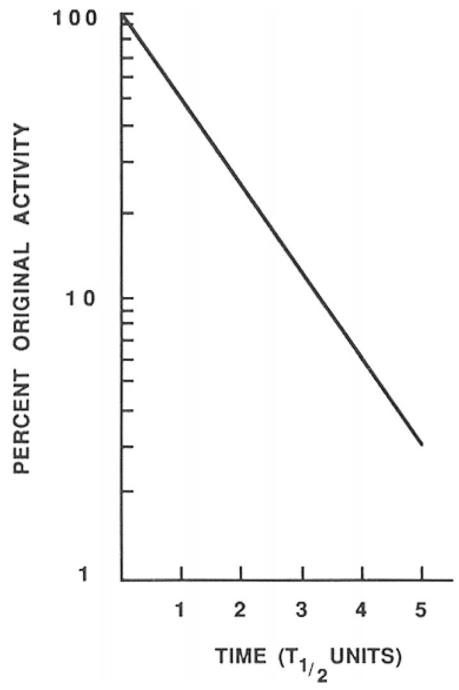
$$T_{1/2} = (\ln 2)/\lambda = 0.693/\lambda$$

Each radioactive isotope has a unique decay constant and, therefore, a unique half-life.

The average life is often described as the *mean life* for radioactive atoms in a sample.



MARGIN FIGURE 1-2A
Percentage of original activity of a radioactive sample as a function of time in units of half-life: Linear plot.



MARGIN FIGURE 1-2B
Percentage of original activity of a radioactive sample as a function of time in units of half-life: Semilogarithmic plot.

where ln 2 is the natural (naperian) logarithm of 2 (logarithm to the base e), and 0.693 is the value of this logarithm. The average life t_{avg} of a radioactive sample, sometimes referred to as the mean life, is the average time for decay of atoms in the sample. The average life is $t_{avg} = 1/\lambda = 1.44(T_{1/2})$.

Example 1-5

What are the half-life and average life of the sample of $^{60}_{27}$Co described in Example 1-4?

$$\begin{aligned} T_{1/2} &= 0.693/\lambda = 0.693/0.131\ \text{y}^{-1} \\ &= 5.3\ \text{y} \\ T_{avg} &= 1.44(T_{1/2}) = 1.44(5.3\ \text{y}) \\ &= 7.63\ \text{y} \end{aligned}$$

The percent of original activity remaining in a radioactive sample is depicted in Margin Figure 1-2A as a function of elapsed time. This variable is replotted in Margin Figure 1-2B on a semilogarithmic graph (activity on a vertical logarithmic scale and time on a horizontal linear scale) to yield a straight line. Semilogarithmic plots yield straight lines of variables such as activity that vary according to an exponential relationship and are useful in depicting several quantities in radiation therapy (e.g., radioactive decay, attenuation of radiation, and survival of tumor cells following irradiation).

Example 1-6

The physical half-life of ^{131}I is 8.0 days.

A. A sample of ^{131}I has a mass of 100 μg. How many ^{131}I atoms are present in the sample?

$$\begin{aligned} &\text{Number of atoms } N \\ &= \frac{(\text{Number of grams})\ (\text{Number of atoms/g-atomic mass})}{(\text{Number of g/g-atomic mass})} \\ &= \frac{(100 \times 10^{-6} g)(6.02 \times 10^{23} \text{atoms/g-atomic mass})}{131\ \text{g/g-atomic mass}} \\ &= 4.6 \times 10^{17} \text{atoms} \end{aligned}$$

B. How many ^{131} I atoms remain after 20 days have elapsed?

$$\begin{aligned} N &= N_0 e^{-(0.693\, t/T_{1/2})} \\ &= (4.6 \times 10^{17}\ \text{atoms}) e^{-(0.693/8\,\text{d})(20\,\text{d})} \\ &= 8.1 \times 10^{16}\ \text{atoms} \end{aligned}$$

C. What is the activity of the sample after 20 days?

$$\begin{aligned} A &= \lambda N \\ &= (0.693/8.0\,\text{d})(1/86,400\ \text{s/d})(8.1 \times 10^{16}\ \text{atoms}) \\ &= 8.2 \times 10^{10}\ \text{atoms/sec} \\ &= 8.2 \times 10^{4}\ \text{MBq} \end{aligned}$$

D. What is the specific activity of the ^{131}I sample?

$$\begin{aligned} \text{SA} &= 8.2 \times 10^{4}\ \text{MBq}/0.1\ \text{mg} \\ &= 8.2 \times 10^{5}\ \text{MBq/mg} \end{aligned}$$

E. What activity should be ordered at 8 AM Monday to provide an activity of 8.2×10^4 MBq at 8 AM on the following Friday?

$$\text{Elapsed time} = 4 \text{ days}$$
$$N = N_0 e^{-\lambda t}$$
$$8.2 \times 10^4 \text{ MBq} = N_0 e^{-(0.693/8\text{d})(4\text{d})}$$
$$8.2 \times 10^4 \text{ MBq} = N_0(0.7072)$$
$$N_0 = 11.6 \times 10^4 \text{ MBq must be ordered}$$

TYPES OF RADIOACTIVE DECAY

The process of radioactive decay often is described by a decay scheme in which energy is depicted on the vertical (y) axis and atomic number is shown on the horizontal (x) axis. A generic decay scheme is illustrated in Figure 1-5. The original nuclide (or "parent") is depicted as ${}^{A}_{Z}X$, and the product nuclide (or "progeny") is denoted as element P, Q, R, or S depending on the decay path. In the path from X to P, the nuclide gains stability by emitting an alpha (α) particle, two neutrons and two protons ejected from the nucleus as a single particle. In this case, the progeny nucleus has an atomic number of $Z - 2$ and a mass number of A$-$4 and is positioned at reduced elevation in the decay scheme to demonstrate that energy is released as the nucleus gains stability through radioactive decay. The released energy is referred to as the *transition energy*. In the path from X to Q, the nucleus gains stability through the process in which a proton in the nucleus changes to a neutron. This process can be either positron decay or electron capture and yields an atomic number of Z $-$1 and an unchanged mass number A. The path from X to R represents negatron decay in which a neutron is transformed into a proton, leaving the progeny with an atomic number of $Z + 1$ and an unchanged mass number A. In the path from R to S, the constant Z and constant A signify that no change occurs in nuclear composition. This pathway is termed an isomeric transition between nuclear isomers and results only in the release of energy from the nucleus through the processes of gamma emission and internal conversion.

A decay scheme is a useful way to assimilate and depict the decay characteristics of a radioactive nuclide.

Parent and progeny nuclei were referred to in the past as "mother" and "daughter." The newer and preferred terminology of parent and progeny is used in this text.

The transition energy released during radioactive decay is also referred to as the "disintegration energy" and the "energy of decay."

Neutrons can be transformed to protons, and vice versa, by rearrangement of their constituent quarks.

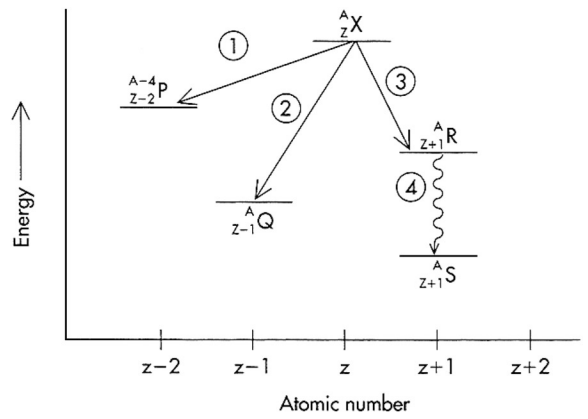


FIGURE 1-5
Symbolic radioactive decay scheme.

Alpha Decay

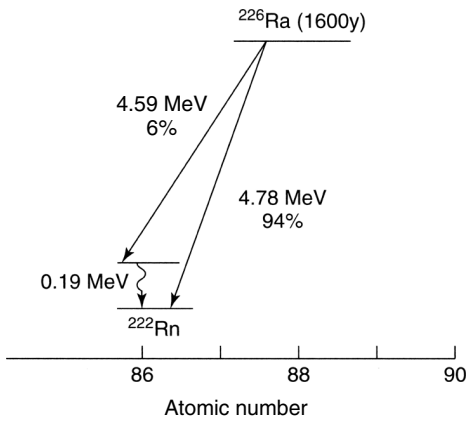
Alpha decay is a decay process in which greater nuclear stability is achieved by emission of 2 protons and 2 neutrons as a single alpha (α) particle (a nucleus of helium) from the nucleus. Alpha emission is confined to relatively heavy nuclei such

Alpha decay was discovered by Marie and Pierre Curie[5] in 1898 in their efforts to isolate radium, and was first described by Ernest Rutherford[6] in 1899. Alpha particles were identified as helium nuclei by Boltwood and Rutherford in 1911.[7] The Curies shared the 1902 Nobel Prize in Physics with Henri Becquerel.

After a lifetime of scientific productivity and two Nobel prizes, Marie Curie died in Paris at the age of 67 from aplastic anemia, probably the result of years of exposure to ionizing radiation.

Rutherford, revered as a teacher and research mentor, was known as "Papa" to his many students.

A radionuclide is a radioactive form of a nuclide.



MARGIN FIGURE 1-3A
Radioactive decay scheme: α decay of ^{226}Ra.

Negative and positive electrons emitted during beta decay are created at the moment of decay. They do not exist in the nucleus before decay.

Ernest Rutherford first characterized beta decay in 1899.[6]

The difference in the energy released during decay, and that possessed by the negatron, threatened the concept of energy conservation for several years. In 1933 Wolfgang Pauli[8] suggested that a second particle was emitted during each decay that accounted for the energy not carried out by the negatron. This particle was named the *neutrino* (Italian for "little neutral particle") by Enrico Fermi.

Enrico Fermi was a physicist of astounding insight and clarity who directed the first sustained man-made nuclear chain reaction on December 2, 1942 in the squash court of the University of Chicago stadium. He was awarded the 1938 Nobel Prize in Physics.

as ^{226}Ra:

$$^{226}_{88}\text{Ra} \rightarrow {}^{222}_{86}\text{Rn} + {}^{4}_{2}\text{He}$$

where $^{4}_{2}$He represents the alpha particle. The sum of mass numbers and the sum of atomic numbers after the transition equal the mass and atomic numbers of the parent before the transition. In α decay, energy is released as kinetic energy of the α particle, and is sometimes followed by energy released during an isomeric transition resulting in emission of a γ ray or conversion electron. Alpha particles are always ejected with energy characteristic of the particular nuclear transition.

An alpha transition is depicted in the margin, in which the parent ^{226}Ra decays directly to the final energy state (ground state) of the progeny ^{222}Rn in 94% of all transitions. In 6% of the transitions, ^{226}Ra decays to an intermediate higher energy state of ^{222}Rn, which then decays to the ground state by isomeric transition. For each of the transition pathways, the transition energy between parent and ground state of the progeny is constant. In the example of ^{226}Ra, the transition energy is 4.78 MeV.

Beta Decay

Nuclei with an N/Z ratio that is above the line of stability tend to decay by a form of beta (β) decay known as negatron emission. In this mode of decay, a neutron is transformed into a proton, and the Z of the nucleus is increased by 1 with no change in A. In this manner, the N/Z ratio is reduced, and the product nucleus is nearer the line of stability. Simultaneously an electron (termed a negative beta particle or negatron) is ejected from the nucleus together with a neutral massless particle, termed a neutrino (actually an "antineutrino" in negatron decay), that carries away the remainder of the released energy that is not accounted for by the negatron. The neutrino (or antineutrino) seldom interacts with matter and is not important to applications of radioactivity in medicine.

The process of negatron emission may be written

$$^{1}_{0}\text{n} \rightarrow {}^{1}_{1}\text{p} + {}^{0}_{-1}\text{e} + \bar{\nu}$$

$$\rightarrow {}^{1}_{1}\text{p} + {}^{0}_{-1}\beta + \bar{\nu}$$

where $^{0}_{-1}e$ depicts the ejected negatron (negative beta particle) and $^{0}_{-1}\beta$ reflects the nuclear origin of the negatron. The symbol $\bar{\nu}$ represents the antineutrino. An example of negatron emission is beta decay of ^{60}Co:

$$^{60}_{27}\text{Co} \rightarrow {}^{60}_{28}\text{Ni} + {}^{0}_{-1}\beta + \bar{\nu} + \text{isomeric transition}$$

with the isomeric transition often accomplished by release of cascading gamma rays of 1.17 and 1.33 MeV. A decay scheme for ^{60}Co is shown in the margin. The transition energy for decay of ^{60}Co is 2.81 MeV.

A discrete amount of energy is released when a negatron is emitted from the nucleus. This energy is depicted as the maximum energy $E_{\max}$ of the negatron. Negatrons, however, usually are emitted with some fraction of this energy, and the remainder is carried from the nucleus by the antineutrino. The mean energy of the negatron is $E_{\max}/3$. An energy spectrum of 0.31 MeV $E_{\max}$ negatrons emitted from ^{60}Co is shown in Figure 1–6. Negatron energy spectra are specific for each negatron transition in every nuclide by this mode of nuclear transformation.

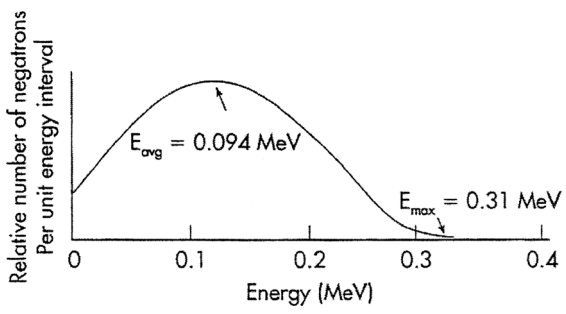


FIGURE 1-6
Energy spectrum of negatrons from $^{60}_{27}$Co.

Example 1-7

Determine the transition energy and the E_{max} of negatrons released during the decay of $^{60}_{27}$Co (atomic mass 59.933814 amu) to $^{60}_{28}$Ni (atomic mass 59.930787 amu).

$$\text{Transition:}\quad {}^{60}_{27}\text{Co}\left[+{}^{0}_{-1}\text{e}\right] \rightarrow {}^{60}_{28}\text{Ni} + {}^{0}_{-1}\beta + \bar{\nu} + \text{isomeric transmission}$$

where the $^{0}_{-1}$e on the left side of the transition must be added from outside the atom to balance the additional positive nuclear charge of ^{60}Ni compared with ^{60}Co.

$$\begin{aligned}\text{Mass difference} &= \text{mass}\left({}^{60}_{27}\text{Co} + {}^{0}_{-1}e\right) - \text{mass}\left({}^{60}_{28}\text{Ni} + {}^{0}_{-1}\beta\right)\\ &= (59.933814 + 0.00055)\,\text{amu} - (59.930787 + 0.00055)\,\text{amu}\\ &= 0.003027\,\text{amu}\\ \text{Transition energy} &= (0.003027\,\text{amu})\,(931\,\text{MeV/amu})\\ &= 2.81\,\text{MeV}\end{aligned}$$

The isomeric transition in ^{60}Co accounts for $(1.17 + 1.33) = 2.50$ MeV (Figure 1-3B). Hence the negatron E_{max} is $2.81 - 2.50 = 0.31$ MeV.

Nuclei below the line of stability are unstable because they have too few neutrons for the number of protons in the nucleus. These nuclei tend to gain stability by a decay process in which a proton is transformed into a neutron, resulting in a unit decrease in Z with no change in A. One possibility for this transformation is positron decay:

$$\begin{aligned}{}^{1}_{1}p &\rightarrow {}^{1}_{0}\text{n} + {}^{0}_{+1}\text{e} + \nu\\ &\rightarrow {}^{1}_{0}\text{n} + {}^{0}_{+1}\beta + \nu\end{aligned}$$

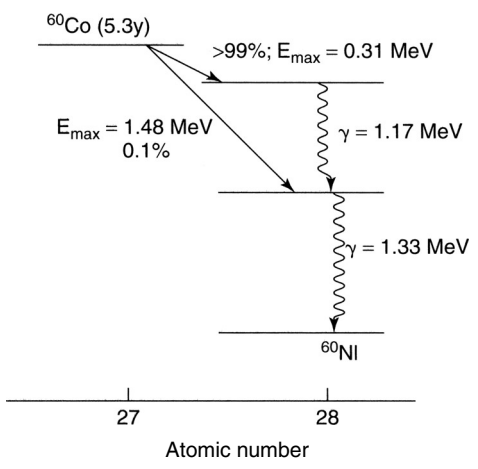
where $^{0}_{+1}\beta$ represents the nuclear origin of the emitted positive electron (positron). A representative positron transition is

$${}^{18}_{9}\text{F} \rightarrow {}^{18}_{8}\text{O} + {}^{0}_{+1}\beta + \nu$$

where ν represents the release of a neutrino, a noninteractive particle similar to an antineutrino except with opposite axial spin. In positron decay, the atomic mass of the decay products exceeds the atomic mass of the atom before decay. This difference in mass must be supplied by energy released during decay according to the relationship $E = \text{mc}^2$. The energy requirement is 1.02 MeV. Hence, nuclei with a transition energy less than 1.02 MeV cannot undergo positron decay. For nuclei with transition energy greater than 1.02 MeV, the energy in excess of 1.02 MeV is shared among the kinetic energy of the positron, the energy of the neutrino, and the energy released during isomeric transitions. Decay of ^{18}F is depicted in the margin, in which the vertical component of the positron decay pathway represents the 1.02 MeV of energy that is expressed as increased mass of the products of the decay process.

An alternate pathway to positron decay is electron capture, in which an electron from an extranuclear shell, usually the K shell, is captured by the nucleus and

The antineutrino was detected experimentally by Reines[9] and Cowan in 1953. They used a 10-ton water-filled detector to detect antineutrinos from a nuclear reactor at Savannah River, SC. Reines shared the 1995 Nobel Prize in Physics.

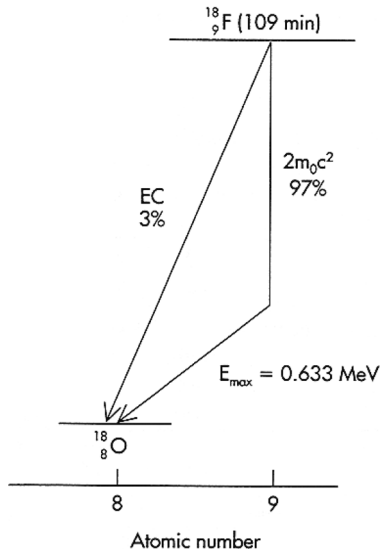


MARGIN FIGURE 1-3B
Radioactive decay scheme: Negatron decay of ^{60}Co.

The emission of positrons from radioactive nuclei was discovered in 1934 by Irene Curie[10] (daughter of Marie Curie) and her husband Frederic Joliet. In bombardments of aluminum by α particles, they documented the following transmutation:

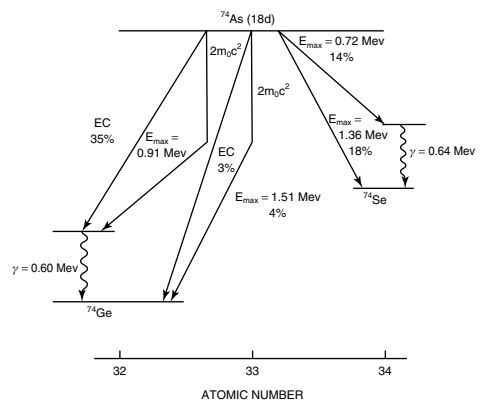
$${}^{4}_{2}\text{He} + {}^{27}_{13}\text{Al} \rightarrow {}^{30}_{15}\text{P} + {}^{1}_{0}\text{n}$$
$${}^{30}_{15}\text{P} \rightarrow {}^{30}_{14}\text{Si} + {}^{0}_{+1}\beta + \upsilon$$

$1.02\ \text{MeV} = 2m_0c^2$, where m_o is the mass of the electron.



MARGIN FIGURE 1-3C
Radioactive decay scheme: $^{0}_{+1}\beta$: e capture decay of $^{18}_{9}F$.

Electron capture of K-shell electrons is known as K-capture; electron capture of L-shell electrons is known as L-capture; and so on.



MARGIN FIGURE 1-3D
Complex decay pattern of $^{74}_{33}$As.

Gamma rays were discovered by the French physicist Paul Villard in 1900.[11] Rutherford and Andrade confirmed in 1912 that γ rays and x rays are similar kinds of radiation.

combined with a proton to transform it into a neutron. The process is represented as

$$^{1}_{1}p + {}^{0}_{1}\mathrm{e} \rightarrow {}^{0}_{1}\mathrm{n} + \nu$$

Electron capture does not yield a mass imbalance before and after the transformation. Hence, there is no transition energy prerequisite for electron capture. Low N/Z nuclei with transition energy less than 1.02 MeV can decay only by electron capture. Low N/Z nuclei with transition energy greater than 1.02 MeV can decay by both positron decay and electron capture. For these nuclei, the electron capture branching ratio describes the probability of electron capture, and (1-branching ratio) depicts the probability of positron decay. Usually, positron decay occurs more frequently than electron capture for nuclei that decay by either process. In the figure illustrating electron capture and positron decay in the margin, the branching ratio for electron capture of ^{18}F is 3%.

Example 1-8

Determine the transition energy and $E_{\max}$ of positrons released during the transformation of $^{18}_{9}$F (atomic mass = 18.000937 amu) to $^{18}_{8}$O (atomic mass = 17.999160 amu). There are no isomeric transitions in this decay process.

$$\text{Transition:} \quad {}^{18}_{9}\mathrm{F} \rightarrow {}^{18}_{8}\mathrm{O} + {}^{0}_{1}\beta + \nu + {}^{0}_{1}\mathrm{e}$$

where the $^{0}_{1}e$ on the right side of the transition must be released from the atom to balance the reduced positive nuclear charge of ^{18}O compared with ^{18}F.

$$\begin{aligned}
\text{Mass difference} &= \text{mass}\left({}^{18}_{9}\mathrm{F}\right) - \text{mass}\left({}^{18}_{8}\mathrm{O} + {}^{0}_{+1}\beta + {}^{0}_{+1}\mathrm{e}\right) \\
&= (18.000937)\,\text{amu} - (17.999160 + 2(0.00055)\,\text{amu} \\
&= 0.000677\,\text{amu} \\
\text{Energy available as } E_{\max} &= (0.000677\,\text{amu})(931\,\text{MeV/amu}) \\
&= 0.630\,\text{MeV}
\end{aligned}$$

The energy equivalent to the mass of the $^{0}_{+1}\beta$ and $^{0}_{1}$e is 2(0.00055 amu) (931 MeV/amu) = 1.02 MeV. Hence the total transition energy is (0.63 + 1.02) MeV = 1.65 MeV.

A few unstable nuclei can decay by negatron decay, positron emission, or electron capture. For example, the decay scheme for ^{74}As in the margin reveals that negatron decay occurs 32% of the time, positron emission occurs with a frequency of 30%, and the nuclide decays by electron capture 38% of the time.

Gamma Emission and Internal Conversion

Frequently during radioactive decay, a product nucleus is formed in an "excited" energy state above the ground energy level. Usually the excited state decays instantly to a lower energy state, often the ground energy level. Occasionally, however, the excited state persists with a finite half-life. An excited energy state that exists for a finite time before decaying is termed a *metastable* energy state and denoted by an m following the mass number (e.g., ^{99m}Tc, which has a half-life of 6 hours). The transition from an excited energy state to one nearer the ground state, or to the ground state itself, is termed an *isomeric transition* because the transition occurs between isomers with no change in Z, N, or A. An isomeric transition can occur by either of two processes: gamma emission or internal conversion.

Gamma rays are high-energy electromagnetic radiation that differ from x rays only in their origin: Gamma rays are emitted during transitions between isomeric energy states of the nucleus, whereas x rays are emitted during electron transitions outside the nucleus. Gamma rays and other electromagnetic radiation are described by their energy E and frequency ν, two properties that are related by the expression $E = h\nu$, where h = Planck's constant ($h = 6.62 \times 10^{-34}$ J-sec). The frequency ν

and wavelength λ of electromagnetic radiation are related by the expression $\nu = c/\lambda$, where c is the speed of light in a vacuum.

No radioactive nuclide decays solely by gamma emission; an isomeric transition is always preceded by a radioactive decay process, such as electron capture or emission of an alpha particle, negatron, or positron. Isomeric transitions for ^{60}Co (as depicted in an earlier marginal figure) yield gamma rays of 1.17 and 1.33 MeV with a frequency of more than 99%. Gamma rays are frequently used in medicine for detection and diagnosis of a variety of ailments, as well as for treatment of cancer.

Internal conversion is a competing process to gamma emission for an isomeric transition between energy states of a nucleus. In a nuclear transition by internal conversion, the released energy is transferred from the nucleus to an inner electron, which is ejected with a kinetic energy equal to the transferred energy reduced by the binding energy of the electron. Internal conversion is accompanied by emission of x rays and Auger electrons as the electron structure of the atom resumes a stable configuration following ejection of the conversion electron. The *internal conversion coefficient* is the fraction of conversion electrons divided by the number of gamma rays emitted during a particular isomeric transition. The conversion coefficient can be expressed in terms of specific electron shells denoting the origin of the conversion electron. The probability of internal conversion increases with Z and the lifetime of the excited state of the nucleus.

RADIOACTIVE EQUILIBRIUM

Some progeny nuclides produced during radioactive decay are themselves unstable and undergo radioactive decay in a continuing quest for stability. For example, ^{226}Ra decays to ^{222}Rn, which, in turn, decays by alpha emission to ^{218}Po. When a radioactive nuclide is produced by radioactive decay of a parent, a condition can be reached in which the rate of production of the progeny equals its rate of decay. In this condition, the number of progeny atoms and therefore the progeny activity reach their highest level and are constant for a moment in time. This constancy reflects an equilibrium condition known as *transient equilibrium* because it exists only momentarily. In cases in which a shorter-lived radioactive progeny is produced by decay of a longer-lived parent, the activity curves for parent and progeny intersect at the moment of transient equilibrium. This intersection reflects the occurrence of equal activities of parent and daughter at that particular moment. After the moment of transient equilibrium has passed, the progeny activity decays with an apparent half-life equal to that of the longer-lived parent. The apparent half-life of the progeny reflects the simultaneous production and decay of the progeny.

If no progeny atoms are present at time $t = 0$, the number N_2 of progeny atoms at any later time t is:

$$N_2 = [\lambda_1/(\lambda_2 - \lambda_1)]N_0(e^{-\lambda_1 t}) \qquad \textbf{(1-5)}$$

In this expression, N_0 is the number of parent atoms present at time $t = 0$, λ_1 is the decay constant of the parent, and λ_2 is the decay constant of the progeny. If $(N_2)_0$ progeny atoms are present at time $t = 0$, the expression for N_2 is written

$$N_2 = (N_2)_0 e^{-\lambda_2 t} + [\lambda_1/(\lambda_2 - \lambda_1)]N_0(e^{-\lambda_1 t} - e^{-\lambda_2 t})$$

Transient equilibrium for a hypothetical nuclide Y formed by decay of the parent X is illustrated in the margin. The activity of Y is greatest at the moment of transient equilibrium and exceeds the activity of X at all times after transient equilibrium is achieved, provided that no amount of Y is removed from the sample. After transient equilibrium, the activity of progeny Y decays with an apparent half-life equal to that of the parent X. The ratio of activities A_1 and A_2 for X and Y, respectively, is

$$A_1/A_2 = (\lambda_2 - \lambda_1)/\lambda_2$$

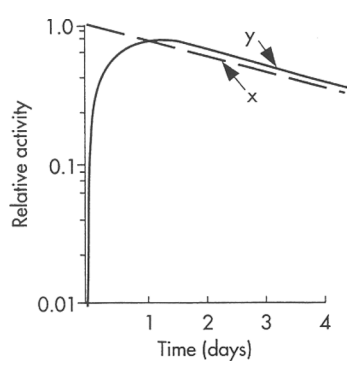
In some texts, transient equilibrium is defined as the extended period over which the progeny decays with an apparent half-life equal to the half-life of the parent. This definition is invalid, because no equilibrium exists beyond the moment when the rate of production of the progeny equals its rate of decay.

Equation (1–5) is a Bateman equation. More complex Bateman equations describe progeny activities for sequential phases of multiple radioactive nuclides in transient equilibrium.

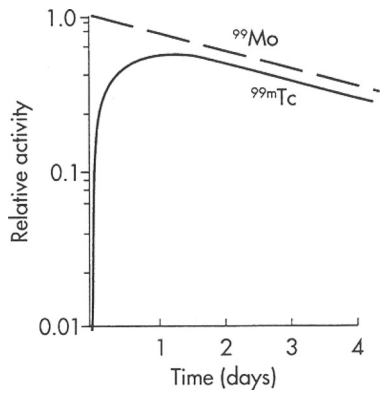
The inert gas ^{222}Rn produced by decay of naturally occurring ^{226}Ra is also radioactive, decaying with a half-life of 3.83 days. This radioactive gas, first called "radium-emanation," was characterized initially by Rutherford. Seepage of ^{222}Rn into homes built in areas with significant ^{226}Ra concentrations in the soil is an ongoing concern to homeowners and the Environmental Protection Agency.

In the hypothetical transient equilibrium between parent X and progeny Y, equilibrium occurs

- at only one instant of time
- when Y reaches its maximum activity
- when the activity of Y is neither increasing or decreasing
- when the activities of X and Y are equal

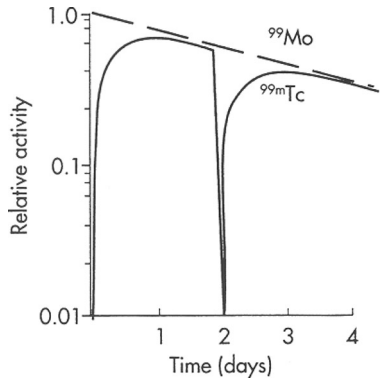


MARGIN FIGURE 1-4A
Transient equilibrium. Hypothetical radionuclide Y formed by decay of parent X.



MARGIN FIGURE 1-4B
Transient equilibrium. Formation of ^{99m}Tc by decay of ^{99}Mo.

The ^{99}Mo–^{99m}Tc generator was developed by Powell Richards in 1957.



MARGIN FIGURE 1-4C
Transient equilibrium. Reestablishment of equilibrium after "milking" a ^{99m}Tc generator.

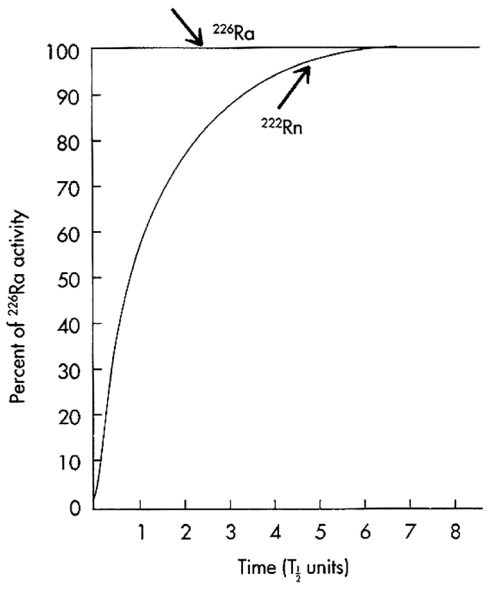


FIGURE 1-7
Growth of activity and secular equilibrium of ^{222}Rn formed by decay of ^{226}Ra.

The principle of transient equilibrium is employed in the production of short-lived nuclides useful in nuclear medicine. The nuclide ^{99m}Tc ($T_{1/2}$ = 6 hours), used in more than 85% of all nuclear medicine examinations, is produced in a radionuclide generator in which the progeny ^{99m}Tc is produced by decay of the parent ^{99}Mo ($T_{1/2}$ = 67 hours). This process is illustrated in the margin, in which the moment of transient equilibrium is illustrated as the point of greatest activity in the curve for ^{99m}Tc. In this case, the ^{99m}Tc activity never reaches that of the parent ^{99}Mo because not all of the ^{99}Mo atoms decay through the isomeric energy state ^{99m}Tc. In a ^{99m}Tc generator, the progeny atoms are removed periodically by "milking the cow" (i.e., removing activity from the generator) by using saline solution to flush an ion exchange column on which the parent is firmly attached. This process gives rise to abrupt decreases in ^{99m}Tc activity, as depicted in the margin.

When the half-life of the parent greatly exceeds that of the progeny (e.g., by a factor of 10^4 or more), equilibrium of the progeny activity is achieved only after a long period of time has elapsed. The activity of the progeny becomes relatively constant, however, as the progeny activity approaches that of the parent, a condition depicted in Figure 1-7. This condition is known as *secular equilibrium* and is a useful approach for the production of the nuclide ^{222}Rn, which was used at one time in radiation therapy. For radionuclides approaching secular equilibrium, the activities of parent (A_1) and progeny (A_2) are equal, and the number of atoms of parent N_1 (which is essentially N_0 because few atoms have decayed since time $t = 0$) and progeny (N_2) are related by the expression

$$A_1 = A_2$$
$$\lambda_1 N_1 = \lambda_2 N_2$$
$$N_0/(T_{1/2})_1 = N_2/(T_{1/2})_2$$

An intraophthalmic irradiator containing ^{90}Sr sometimes is used to treat various conditions of the eye. The low-energy beta particles from ^{90}Sr are not useful clinically, but the higher-energy beta particles from the progeny ^{90}Y are useful. The relatively short-lived Y ($T_{1/2}$ = 64 hours) is contained in the irradiator in secular equilibrium with the longer-lived parent ^{90}Sr ($T_{1/2}$ = 28 years) so that the irradiator can be used over many years without replacement. Radium needles and capsules that were formerly used widely in radiation oncology contained many decay products in secular equilibrium with the long-lived ($T_{1/2}$ = 1600 years) parent ^{226}Ra.

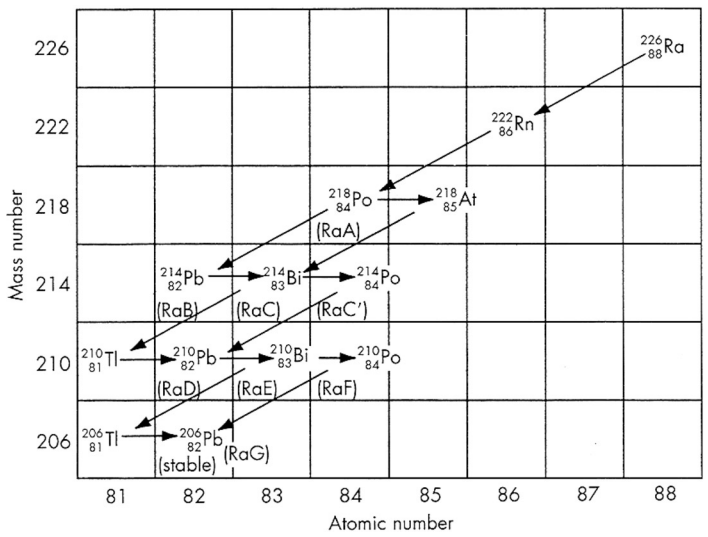


FIGURE 1-8
Uranium ($4n = 2$) decay series.

NATURAL RADIOACTIVITY AND DECAY SERIES

Most radionuclides in nature are members of one of three naturally occurring radioactive decay series. Each series consists of a sequence of radioactive transformations that begins with a long-lived radioactive parent and ends with a stable nuclide. In a closed environment such as the earth, intermediate radioactive progeny exist in secular equilibrium with the long-lived parent, and decay with an apparent half-life equal to that of the parent. All naturally occurring radioactive nuclides decay by emitting either alpha or negative beta particles. Hence, each transformation in a radioactive series changes the mass number by either 4 or 0 and changes the atomic number by -2 or $+1$.

The uranium series depicted in Figure 1-8 begins with the isotope ^{238}U and ends with the stable nuclide ^{206}Pb. The parent and each product in this series have a mass number that is divisible by 4 with remainder of 2; the uranium series is also known as the $4n + 2$ series. The naturally occurring isotopes ^{226}Ra and ^{222}Rn are members of the uranium series. The actinium ($4n + 3$) series begins with ^{235}U and ends with ^{207}Pb, and the thorium ($4n$) series begins with ^{232}Th and ends with ^{208}Pb. Members of the hypothetical neptunium ($4n + 1$) series do not occur in nature because no long-lived parent is available. Fourteen naturally occurring radioactive nuclides are not members of a decay series. These nuclides, all with relatively long half-lives, are ^{3}H, ^{14}C, ^{40}K, ^{50}V, ^{87}Rb, ^{115}In, ^{130}Te, ^{138}La, ^{142}Ce, ^{144}Nd, ^{147}Sm, ^{176}Lu, ^{187}Re, and ^{192}Pt.

ARTIFICIAL PRODUCTION OF RADIONUCLIDES

Radioactive isotopes with properties useful in biomedical research and clinical medicine may be produced by bombarding selected nuclei with neutrons and high-energy charged particles. Nuclides with excess neutrons that subsequently decay by negatron emission are created by bombarding nuclei with neutrons in a nuclear reactor or from a neutron generator. Typical reactions are

$$^{13}_{6}\mathrm{C} + ^{1}_{0}\mathrm{n} \rightarrow ^{14}_{6}\mathrm{C} + \text{isomeric transition}$$

$$^{31}_{15}\mathrm{P} + ^{1}_{0}\mathrm{n} \rightarrow ^{32}_{15}\mathrm{P} + \text{isomeric transition}$$

Useful isotopes produced by neutron bombardment include ^{3}H, ^{35}S, ^{51}Cr, ^{60}Co,

The first high-energy particle accelerator was the cyclotron developed by Ernest Lawrence in 1931. In 1938, Ernest and his physician brother John used artificially produced ^{32}P to treat their mother who was afflicted with leukemia.

^{99}Mo, ^{133}Xe, and ^{198}Au. Because the isomeric transition frequently results in prompt emission of a gamma ray, neutron bombardment often is referred to as an (n, γ) reaction. The reaction yields a product nuclide with an increase in A of 1 and no increase in Z. The complete transformation, including radioactive decay that results from neutron bombardment, is demonstrated by the example of ^{60}Co:

$$^{59}_{27}\text{Co} + {}^{1}\text{n} \rightarrow {}^{60}_{27}\text{Co} + \text{isomeric transitions}$$

$$^{60}_{27}\text{Co} \rightarrow {}^{60}_{28}\text{Ni} + {}^{0}_{-1}\beta + \bar{\nu} + \text{isomeric transitions}$$

The transition can be represented as ^{59}Co$(n, \gamma)^{60}$Co. The decay of ^{60}Co occurs with a half-life of 5.3 years. The isomeric transitions accompanying this decay process almost always result in emission of cascading gamma rays of 1.17 and 1.33 MeV.

Radionuclides with excess protons are produced when nuclei are bombarded with high-energy positively charged particles from a particle accelerator. These radionuclides then decay by electron capture and, if the transition energy is adequate, positron decay. A typical reaction is

$$^{11}_{5}\text{B} + {}^{1}_{1}\text{P} \rightarrow {}^{11}_{6}\text{C} + {}^{1}_{0}\text{n}$$

where $^{1}_{0}$n denotes that a neutron is ejected from the nucleus during bombardment so that the parent and progeny nuclei are isobars. This reaction can be represented as ^{11}B(p,n)^{11}C and is termed a (p,n) reaction. Other representative charged-particle interactions include

$$^{14}_{7}\text{N} + {}^{4}_{2}\text{He} \rightarrow {}^{17}_{8}\text{O} + {}^{1}_{1}\text{p} \qquad \text{(an } (\alpha, \text{p}) \text{ reaction)}$$

$$^{68}_{30}\text{Zn} + {}^{1}_{1}\text{p} \rightarrow {}^{67}_{31}\text{Ga} + 2{}^{1}_{0}\text{n} \qquad \text{(a (p, 2n) reaction)}$$

$$^{27}_{13}\text{Al} + {}^{4}_{2}\text{He} \rightarrow {}^{30}_{15}\text{P} + {}^{1}_{0}\text{n} \qquad \text{(an } (\alpha, \text{n}) \text{ reaction)}$$

$$^{12}_{6}\text{C} + {}^{1}_{1}\text{p} \rightarrow {}^{13}_{7}\text{N} + \gamma \qquad \text{(a (p, } \gamma \text{) reaction)}$$

$$^{3}_{1}\text{H} + {}^{2}_{1}\text{d} \rightarrow {}^{4}_{2}\text{He} + {}^{1}_{0}\text{n} \qquad \text{(a (d, n) reaction)}$$

where d stands for deuteron, a particle composed of a proton and neutron (i.e., a nucleus of deuterium).

Radioactive nuclides are also produced as a result of nuclear fission. These nuclides can be recovered as fission byproducts from the fuel elements used in nuclear reactors. Isotopes such as ^{90}Sr, ^{99}Mo, ^{131}I, and ^{137}Cs can be recovered in this manner.

Fission-produced nuclides (fission byproducts) are often mixed with other stable and radioactive isotopes of the same element, and cannot be separated chemically as a solitary radionuclide.[13] As a consequence, fission byproducts are less useful in research and clinical medicine than are radionuclides that are produced by neutron or charged-particle bombardment.

Nuclear transmutation by particle bombardment was first observed by Rutherford[12] in 1919 in his studies of α particles traversing an air-filled chamber. The observed transmutation was

$$^{4}_{2}\text{He} + {}^{14}_{7}\text{N} \rightarrow {}^{17}_{8}\text{O} + {}^{1}_{1}\text{H}$$

where $^{4}_{2}$He represents α particles and $^{1}_{1}$H depicts protons detected during the experiment. This reaction can be written more concisely as

$$^{14}_{7}\text{N}\ (\alpha,\ \text{p})^{17}_{8}\text{O}$$

where $^{14}_{7}N$ represents the bombarded nucleus, $^{17}_{8}$O the product nucleus, and (α, p) the incident and ejected particles, respectively.

Through their discovery of artificial radioactivity, Irene Curie (the daughter of Marie Curie) and Frederic Joliot paved the way to use of radioactive tracers in biomedical research and clinical medicine.

SUMMARY

- Radioactive decay is the consequence of nuclear instability.
  - Negatron decay occurs in nuclei with a high n/p ratio.
  - Positron decay and electron capture occur in nuclei with a low n/p ratio.
  - Alpha decay occurs with heavy unstable nuclei.
  - Isomeric transitions occur between different energy states of nuclei and result in the emission of γ rays and conversion electrons.
- The activity A of a sample is
$$A = A_o e^{-\lambda t}$$
where λ is the decay constant (fractional rate of decay).
- The half-life $T_{1/2}$ is the time required for half of a radioactive sample to decay. The half-life and the decay constant are related by
$$T_{1/2} = 0.693/\lambda$$

- The common unit of activity is the becquerel (Bq), with 1 Bq = 1 disintegration/second.
- Transient equilibrium may exist when the progeny nuclide decays with a $T_{1/2}$ < $T_{1/2}$ parent.
- Secular equilibrium may exist when the progeny nuclide decays with a $T_{1/2} \ll T_{1/2}$ parent.
- Most radioactive nuclides found in nature are members of naturally occurring decay series.

PROBLEMS

1-1. What are the atomic and mass numbers of the oxygen isotope with 17 nucleons? Calculate the mass defect, binding energy, and binding energy per nucleon for this nuclide, with the assumption that the mass defect is associated with the nucleus. The mass of the atom is 16.999133 amu.

1-2. Natural oxygen contains three isotopes with atomic masses in amu of 15.9949, 16.9991, and 17.9992 and relative abundances of 2500:1:5. Determine to three decimal places the average atomic mass of oxygen.

1-3. Determine the energy required to move an electron from the K to the L shell in tungsten and in hydrogen, and explain the difference.

1-4. What is the energy equivalent to the mass of an electron? a proton?

1-5. The energy released during the nuclear explosion at Hiroshima has been estimated as equivalent to that released by 20,000 tons of TNT. Assume that 200 MeV is released when a ^{235}U nucleus absorbs a neutron and fissions and that 3.8×10^9J is released during detonation of 1 ton of TNT. How many nuclear fissions occurred at Hiroshima, and what was the total decrease in mass?

1-6. Group the following nuclides as isotopes, isotones, and isobars:

$$^{14}_{6}\text{C},\ ^{14}_{7}\text{N},\ ^{15}_{7}\text{N},\ ^{15}_{6}\text{C},\ ^{16}_{7}\text{N},\ ^{16}_{8}\text{O},\ ^{17}_{8}\text{O}$$

1-7. The half-life of ^{32}P is 14.3 days. What interval of time is required for 100 mCi of ^{32}P to decay to 25 mCi? What time is required for decay of 7/8 of the ^{32}P atoms?

1-8. A radioactive needle contains $^{222}_{86}$Rn ($T_{1/2}$ = 3.83 days) in secular equilibrium with $^{226}_{88}$Ra ($T_{1/2}$ = 1600 years). How long is required for the $^{222}_{86}$Rn to decay to half of its original activity?

1-9. In nature, $^{226}_{88}$Ra ($T_{1/2}$ = 1600 years) exists in secular equilibrium with $^{238}_{92}$U ($T_{1/2} = 4.5 \times 10^9$ years). What fraction of the world's supply of radium will be left after 1600 years?

1-10. What is the mass in grams of 100 MBq of pure ^{32}P? How many ^{32}P atoms constitute 100 MBq? What is the mass in grams of 100 MBq of Na_3PO_4 if all the phosphorus in the compound is radioactive?

1-11. If a radionuclide decays for an interval of time equal to its average life, what percentage of the original activity remains?

1-12. What are the wavelength and frequency of a 1-MeV photon?

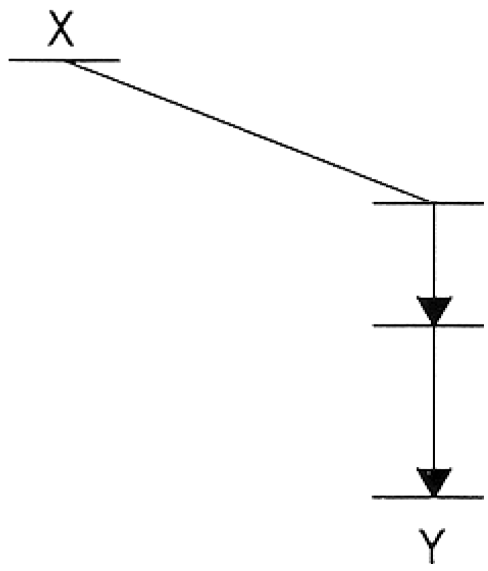
1-13. ^{126}I nuclei decay by negatron emission, positron emission, and electron capture. Write the decay equation for each mode of decay and identify the daughter nuclide.

1-14. How many atoms and grams of ^{90}Y are in secular equilibrium with 50 mCi of ^{90}Sr?

1-15. How many MBq of ^{132}I ($T_{1/2}$ = 2.3 hours) should be ordered so that the sample activity will be 500 MBq when it arrives 24 hours later?

1-16. $^{127}_{53}I$ is the only stable isotope of iodine. What mode(s) of decay would be expected for ^{131}I? ^{125}I?

1-17. For a nuclide X with the decay scheme



how many gamma rays are emitted per 100 disintegrations of X if the coefficient for interval conversion is 0.25?

1-18. ^{3}H (3.016050 amu) decays to $^{3}_{2}$He (3.016030 amu) by negatron emission. What is the transition energy and negatron E_{max} if no isomeric transitions occur?

1-19. $^{11}_{6}$C (11.011432 amu) decays to $^{11}_{5}$B (11.009305 amu) by positron emission and electron capture. What is the transition energy and positron E_{max} if no isomeric transitions occur?

P1: IML/FFX P2: IML/FFX QC: IML/FFX T1: IML
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