Decay of Radioactivity

Radioactive decay is a spontaneous process; that is, there is no way to predict with certainty the exact moment at which an unstable nucleus will undergo its radioactive transformation into another, more stable nucleus. Mathematically, radioactive decay is described in terms of probabilities and average decay rates. In this chapter we discuss these mathematical aspects of radioactive decay.

A. ACTIVITY

1. The Decay Constant

If one has a sample containing N radioactive atoms of a certain radionuclide, the average decay rate, $\Delta N/\Delta t$, for that sample is given by:

$$\Delta N/\Delta t = -\lambda N \qquad (4\text{-}1)$$

where λ is the *decay constant* for the radionuclide. The decay constant has a characteristic value for each radionuclide. It is the fraction of the atoms in a sample of that radionuclide undergoing radioactive decay per unit of time during a period that is so short that only a small fraction decay during that interval. Alternatively, it is the probability that any individual atom will undergo decay during the same period. The units of λ are (time)$^{-1}$. Thus 0.01 sec^{-1} means that, on the average, 1% of the atoms undergo radioactive decay each second. In Equation 4-1 the minus sign indicates that $\Delta N/\Delta t$ is negative; that is, N is decreasing with time.

Equation 4-1 is valid only as an estimate of the *average* rate of decay for a radioactive sample. From one moment to the next, the actual decay rate may differ from that predicted by Equation 4-1. These *statistical fluctuations* in decay rate are described in Chapter 9.

Some radionuclides can undergo more than one type of radioactive decay (e.g., ^{18}F: 97% β^+, 3% electron capture). For such types of "branching" decay, one can define a value of λ for each of the possible decay modes, for example, λ_1, λ_2, λ_3, and so on, where λ_1 is the fraction decaying per unit time by decay mode 1, λ_2 by decay mode 2, and so on. The total decay constant for the radionuclide is the sum of the branching decay constants:

$$\lambda = \lambda_1 + \lambda_2 + \lambda_3 + \cdots \qquad (4\text{-}2)$$

The fraction of nuclei decaying by a specific decay mode is called the *branching ratio* (B.R.). For the ith decay mode, it is given by:

$$\text{B.R.} = \lambda_i/\lambda \qquad (4\text{-}3)$$

2. Definition and Units of Activity

The quantity $\Delta N/\Delta t$, the average decay rate, is the *activity* of the sample. It has dimensions of disintegrations per second (dps) or disintegrations per minute (dpm) and is essentially a measure of "how radioactive" the sample is. The Systeme International (SI) unit of activity is the *becquerel* (Bq). A sample has an activity of 1 Bq if it is decaying at an average rate of 1 sec^{-1} (1 dps). Thus:

$$A(\text{Bq}) = |\Delta N/\Delta t| = \lambda N \qquad (4\text{-}4)$$

where λ is in units of sec^{-1}. The absolute value is used to indicate that activity is a "positive" quantity, as compared with the change in number of radioactive atoms in Equation 4-1, which is a negative quantity. Commonly used multiples of the becquerel are the kilobecquerel (1 kBq = 10^3 sec^{-1}), the megabecquerel (1 MBq = 10^6 sec^{-1}), and the gigabecquerel (1 GBq = 10^9 sec^{-1}).

The traditional unit for activity is the *curie* (Ci), which is defined as 3.7×10^{10} dps (2.22×10^{12} dpm). Subunits and multiples of the curie are the millicurie (1 mCi = 10^{-3} Ci), the microcurie (1 μCi = 10^{-3} mCi = 10^{-6} Ci), the nanocurie (1 nCi = 10^{-9} Ci), and the kilocurie (1 kCi = 1000 Ci). Equation 4-1 may be modified for these units of activity:

$$A(\text{Ci}) = \lambda N/(3.7 \times 10^{10}) \qquad (4\text{-}5)$$

The curie was defined originally as the activity of 1 g of ^{226}Ra; however, this value "changed" from time to time as more accurate measurements of the ^{226}Ra decay rate were obtained. For this reason, the ^{226}Ra standard was abandoned in favor of a fixed value of 3.7×10^{10} dps. This is not too different from the currently accepted value for ^{226}Ra (3.656×10^{10} dps/g).

SI units are the "official language" for nuclear medicine and are used in this text; however, because traditional units of activity still are used in day-to-day practice in many laboratories, we sometimes also indicate activities in these units as well. Conversion factors between traditional and SI units are provided in Appendix A.
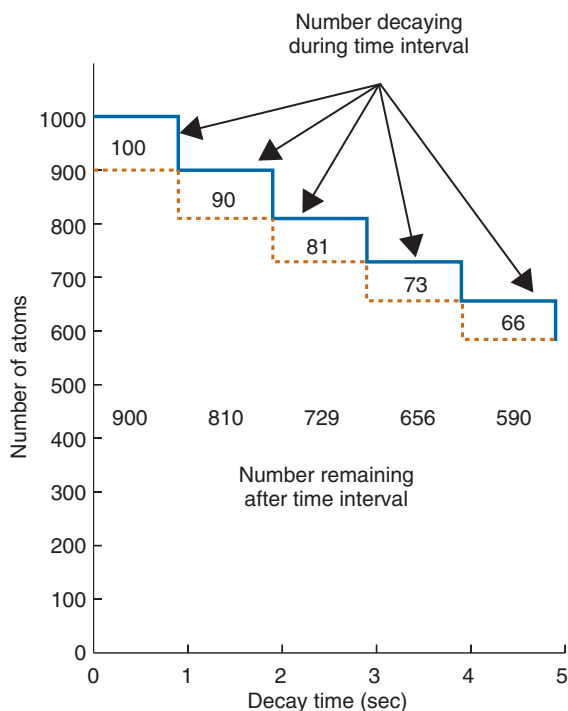
The amounts of activity used for nuclear medicine studies typically are in the MBq-GBq range (10s of μCi to 10s of mCi). Occasionally, 10s of gigabecquerels (curie quantities) may be acquired for long-term supplies. External-beam radiation sources (e.g., ^{60}Co therapy units) use source strengths of 1000s of GBq [1000 GBq = 1 terraBq (TBq) = 10^{12} Bq]. At the other extreme, the most sensitive measuring systems used in nuclear medicine can detect activities at the level of a few becquerels (nanocuries).

B. EXPONENTIAL DECAY

1. The Decay Factor

With the passage of time, the number N of radioactive atoms in a sample decreases. Therefore the activity A of the sample also decreases (see Equation 4-4). Figure 4-1 is used to illustrate radioactive decay with the passage of time.

Suppose one starts with a sample containing $N(0)$ = 1000 atoms* of a radionuclide having a decay constant λ = 0.1 sec^{-1}. During the first 1-sec time interval, the approximate

*$N(t)$ is symbolic notation for the number of atoms present as a function of time t. $N(0)$ is the number N at a specific time t = 0, that is, at the starting point.

FIGURE 4-1 Decay of a radioactive sample during successive 1-sec increments of time, starting with 1000 atoms, for λ = 0.1 sec^{-1}. Both the number of atoms remaining and activity (decay rate) decrease with time. Note that the values shown are approximations, because they do not account precisely for the changing number of atoms present during the decay intervals (see Section D).

number of atoms decaying is 0.1×1000 = 100 atoms (see Equation 4-1). The activity is therefore 100 Bq, and after 1 sec there are 900 radioactive atoms remaining. During the next second, the activity is 0.1×900 = 90 Bq, and after 2 sec, 810 radioactive atoms remain. During the next second the activity is 81 Bq, and after 3 sec 729 radioactive atoms remain. Thus both the activity and the number of radioactive atoms remaining in the sample are decreasing continuously with time. A graph of either of these quantities is a curve that gradually approaches zero.

An exact mathematical expression for $N(t)$ can be derived using methods of calculus.* The result is:

$$N(t) = N(0)\, e^{-\lambda t} \qquad (4\text{-}6)$$

---

*The derivation is as follows:

$$\text{dN/dt} = -\lambda\text{N} \qquad (4\text{-}6a)$$

$$\text{dN/N} = -\lambda\text{dt} \qquad (4\text{-}6b)$$

$$\int d\text{N/N} = -\int \lambda\, dt \qquad (4\text{-}6c)$$

from which follows Equation 4-6.

Thus $N(t)$, the number of atoms remaining after a time t, is equal to $N(0)$, the number of atoms at time $t = 0$, multiplied by the factor $e^{-\lambda t}$. This factor $e^{-\lambda t}$, the fraction of radioactive atoms remaining after a time t, is called the *decay factor* (DF). It is a *number* equal to e—the base of natural logarithms $(2.718 \ldots)$—raised to the power $-\lambda t$. For given values of λ and t, the decay factor can be determined by various methods as described in Section C later in this chapter. Note that because activity A is proportional to the number of atoms N (see Equation 4-4), the decay factor also applies to activity versus time:

$$A(t) = A(0) \, e^{-\lambda t} \qquad (4\text{-}7)$$

The decay factor $e^{-\lambda t}$ is an *exponential function* of time t. Exponential decay is characterized by the disappearance of a *constant fraction* of activity or number of atoms present per unit time interval. For example if $\lambda = 0.1 \text{ sec}^{-1}$, the fraction is 10% per second. Graphs of $e^{-\lambda t}$ versus time t for $\lambda = 0.1 \text{ sec}^{-1}$ are shown in Figure 4-2. On a *linear* plot, it is a curve gradually approaching zero; on a *semilogarithmic* plot, it is a straight line. It should be noted that there are other processes besides radioactive decay that can be described by exponential functions. Examples are the absorption of x- and λ-ray beams (see Chapter 6, Section D) and the clearance of certain tracers from organs by physiologic processes (see Chapter 22, Section B.2).

When the exponent in the decay factor is "small," that is, $\lambda t \lesssim 0.1$, the decay factor may be approximated by $e^{-\lambda t} \approx 1 - \lambda t$. This form may be used as an approximation in Equations 4-6 and 4-7.

2. Half-Life

As indicated in the preceding section, radioactive decay is characterized by the disappearance of a constant fraction of the activity present in the sample during a given time interval. The *half-life* $(T_{1/2})$ of a radionuclide is the time required for it to decay to 50% of its initial activity level. The half-life and decay constant of a radionuclide are related as[*]

$$T_{1/2} = \ln 2/\lambda \qquad (4\text{-}8)$$
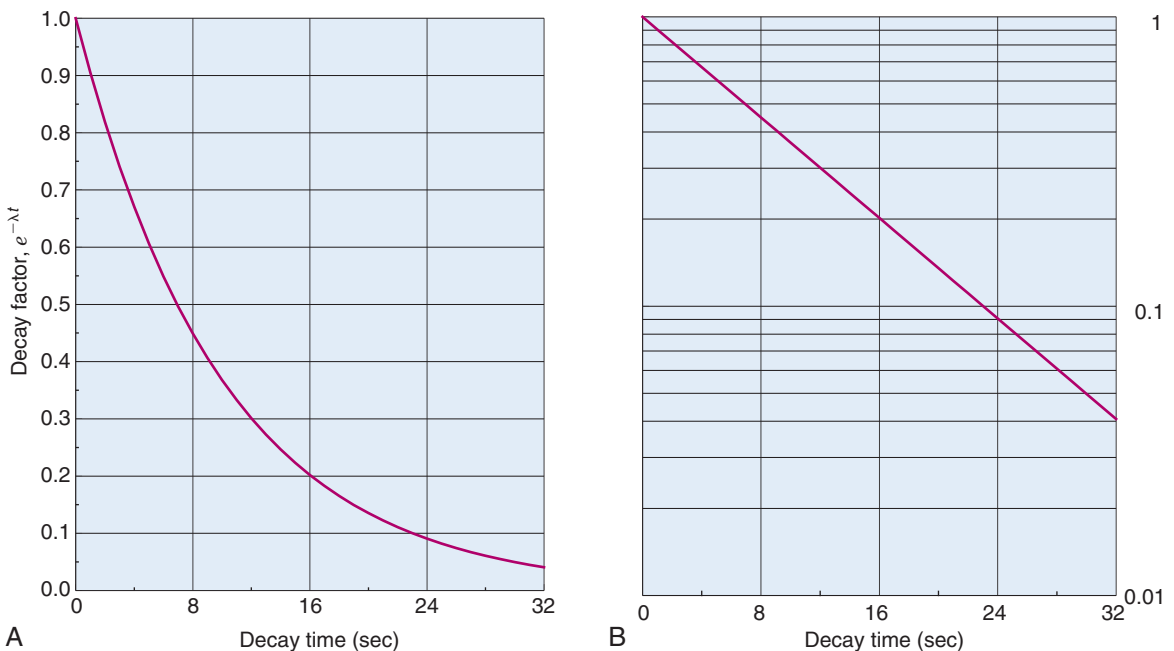
$$\lambda = \ln 2/T_{1/2} \qquad (4\text{-}9)$$

[*]The relationships are derived as follows:

$$1/2 = e^{-\lambda T_{1/2}} \qquad (4\text{-}8a)$$

$$2 = e^{\lambda T_{1/2}} \qquad (4\text{-}8b)$$

$$\ln 2 = \lambda T_{1/2} \qquad (4\text{-}8c)$$

from which follow Equations 4-8 and 4-9.

FIGURE 4-2 Decay factor versus time shown on linear (*A*) and semilogarithmic (*B*) plots, for radionuclide with $\lambda = 0.1 \text{ sec}^{-1}$.

where $\ln 2 \approx 0.693$. Usually, tables or charts of radionuclides list the half-life of the radionuclide rather than its decay constant. Thus it often is more convenient to write the decay factor in terms of half-life rather than decay constant:

$$\mathrm{DF} = e^{-\ln 2 \times t/T_{1/2}} \qquad (4\text{-}10)$$

3. Average Lifetime

The actual lifetimes of individual radioactive atoms in a sample range anywhere from "very short" to "very long." Some atoms decay almost immediately, whereas a few do not decay for a relatively long time (see Fig. 4-2). The *average lifetime* τ of the atoms in a sample has a value that is characteristic of the nuclide and is related to the decay constant λ by*

$$\tau = 1/\lambda \qquad (4\text{-}11)$$

Combining Equations 4-9 and 4-11, one obtains

$$\tau = T_{1/2}/\ln 2 \qquad (4\text{-}12)$$

The average lifetime for the atoms of a radionuclide is therefore longer than its half-life, by a factor $1/\ln 2$ (≈ 1.44). The concept of average lifetime is of importance in radiation dosimetry calculations (see Chapter 22).

C. METHODS FOR DETERMINING DECAY FACTORS

1. Tables of Decay Factors

It is essential that an individual working with radionuclides know how to determine decay factors. Perhaps the simplest and most straightforward approach is to use tables of decay factors, which are available from vendors of radiopharmaceuticals, instrument manufacturers, and so forth. An example of such a table for ^{99m}Tc is shown in Table 4-1. Such tables are generated easily with computer spreadsheet programs.

EXAMPLE 4-1

A vial containing ^{99m}Tc is labeled "75 kBq/mL at 8 am." What volume should be withdrawn

---

*The equation from which Equation 4-11 is derived is:

$$\tau = \int_0^\infty t e^{-\lambda t} dt \bigg/ \int_0^\infty e^{-\lambda t} dt \qquad (4\text{-}11a)$$

TABLE 4-1
DECAY FACTORS FOR ^{99m}Tc

Hours	Minutes			
	0	**15**	**30**	**45**
0	1.000	0.972	0.944	0.917
1	0.891	0.866	0.841	0.817
2	0.794	0.771	0.749	0.727
3	0.707	0.687	0.667	0.648
4	0.630	0.612	0.595	0.578
5	0.561	0.545	0.530	0.515
6	0.500	0.486	0.472	0.459
7	0.445	0.433	0.420	0.408
8	0.397	0.385	0.375	0.364
9	0.354	0.343	0.334	0.324
10	0.315	0.306	0.297	0.289
11	0.281	0.273	0.264	0.257
12	0.250	0.243	0.236	0.229

at 4 pm on the same day to prepare an injection of 50 kBq for a patient?

Answer
From Table 4-1 the DF for ^{99m}Tc after 8 hours is found to be 0.397. Therefore the concentration of activity in the vial is 0.397×75 kBq/mL = 29.8 kBq/mL. The volume required for 50 kBq is 50 kBq divided by 29.8 kBq/mL = 1.68 mL.

Tables of decay factors cover only limited periods; however, they can be extended by employing principles based on the properties of exponential functions, specifically $e^{a+b} = e^a \times e^b$. For example, suppose that the desired time t does not appear in the table but that it can be expressed as a sum of times, $t = t_1 + t_2 + \cdots$, that do appear in the table. Then

$$\mathrm{DF}(t_1 + t_2 + \cdots) = \mathrm{DF}(t_1) \times \mathrm{DF}(t_2) \cdots \quad (4\text{-}13)$$

EXAMPLE 4-2

What is the decay factor for ^{99m}Tc after 16 hours?

Answer
Express 16 hours as 6 hours + 10 hours. Then, from Table 4-1, DF(16 hr) = DF(10 hr) × DF(6 hr) = $0.315 \times 0.5 = 0.1575$. Other combinations of times totaling 16 hours provide the same result.

Occasionally, radionuclides are shipped in *precalibrated* quantities. A precalibrated shipment is one for which the activity calibration is given for some *future* time. To determine its present activity, it is therefore necessary to calculate the decay factor for a time preceding the calibration time, that is, a "negative" value of time. One can make use of tables of decay factors by employing another of the properties of exponential functions, specifically $e^{-x} = 1/e^x$. Thus:

$$DF(-t) = 1/DF(t) \qquad (4\text{-}14)$$

EXAMPLE 4-3

A vial containing ^{99m}Tc is labeled "50 kBq at 3 pm." What is the activity at 8 am on the same day?

Answer
The decay time is $t = -7$ hours. From Table 4-1, DF(7 hr) = 0.445. Thus DF(–7 hr) = 1/0.445 = 2.247. The activity at 8 am is therefore 2.247×50 kBq = 112.4 kBq.

2. Pocket Calculators

Many pocket calculators have capabilities for calculating exponential functions. First compute the exponent, $x = \ln 2 \times (t/T_{1/2})$, then press the appropriate keys to obtain e^{-x}. For precalibrated shipments, use e^{+x}.

3. Universal Decay Curve

Exponential functions are straight lines on a semilogarithmic plot (see Fig. 4-2). This useful property allows one to construct a "universal decay curve" by plotting the number of half-lives elapsed on the horizontal (linear) axis and the decay factor on the vertical (logarithmic) axis. A straight line can be drawn by connecting any two points on the curve. These could be, for example, ($t = 0$, DF = 1), ($t = T_{1/2}$, DF = 0.5), ($t = 2T_{1/2}$, DF = 0.25), and so on. The graph can be used for any radionuclide provided that the elapsed time is expressed in terms of the number of radionuclide half-lives elapsed. An example of a universal decay curve is shown in Figure 4-3.

EXAMPLE 4-4

Use the decay curve in Figure 4-3 to determine the decay factor for ^{99m}Tc after 8 hours.

Answer
The half-life of ^{99m}Tc is 6 hours. Therefore the elapsed time is 8/6 = 1.33 half-lives. From Figure 4-3, the decay factor is approximately
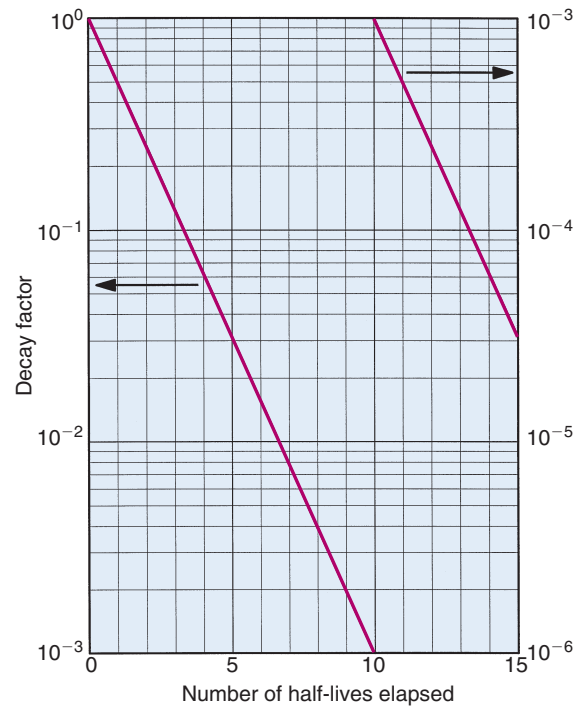

**FIGURE 4-3**   Universal decay curve.

0.40. (Compare this result with the value used in Example 4-1.)

D. IMAGE-FRAME DECAY CORRECTIONS

In some applications, data are acquired during periods that are not short in comparison with the half-life of the radionuclide. An example is the measurement of glucose metabolism using deoxyglucose labeled with fluorine-18 (see Chapter 21, Section E.5). In such measurements, it often is necessary to correct for decay that occurs during each measurement period while data collection is in progress. Because data are acquired in a series of image frames, these sometimes are called *image-frame decay corrections*.

The concept for these corrections is illustrated in Figure 4-4, showing the decay curve for an image frame starting at time t and ending at a time Δt later. The number of counts acquired during the image frame is proportional to the area a_d, shown with darker shading. The counts that would be recorded in the absence of decay are proportional to the area a_0, which includes both the darker and lighter shaded areas. Using the appropriate mathematical integrals, the *effective decay*
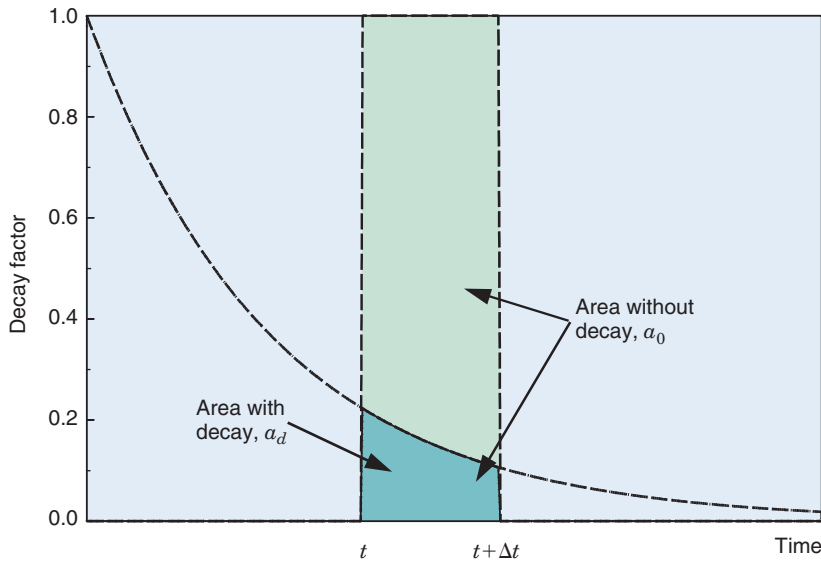
FIGURE 4-4   Basic concept for calculating the decay factor for an image frame starting at time t with duration Δt. The counts recorded with decay are proportional to the darker shaded area, a_d. The counts that would be recorded in the absence of decay are proportional to the total shaded area, a_0. The effective decay factor is the ratio a_d/a_0.

factor for a radionuclide with half-life $T_{1/2}$ for the indicated measurement interval is given by:

$$
\begin{aligned}
\mathrm{DF}_{\mathrm{eff}}\ (t, \Delta t) &= a_d / a_0 \\
&= e^{-(\ln 2 \times t / T_{1/2})} \times [(1 - e^{-x})/x] \\
&= \mathrm{DF}(t) \times [(1 - e^{-x})/x] \quad (4\text{-}15)
\end{aligned}
$$

where

$$
x = \ln 2 \times \Delta t / T_{1/2} \quad (4\text{-}16)
$$

To correct the recorded counts back to what would have been recorded in the absence of decay, one would multiply the counts recorded during the interval $(t, t + \Delta t)$ by the inverse of $\mathrm{DF}_{\mathrm{eff}}$.

The effective decay factor in Equation 4-15 is composed of two parts. The first term is just the standard decay factor (Equation 4-10) at the start of the image frame, $\mathrm{DF}(t)$. The second term is a factor that depends on the parameter x, which in turn depends on the duration of the frame, Δt, relative to the half-life of the radionuclide (Equation 4-16). This term accounts for decay that occurs while data are being acquired during the image frame. Note again that the correction in Equation 4-15 uses $t = 0$ as the reference point, not the start of the individual image frame for which the correction is being calculated. To compute the decay occurring during the image frame itself, only the second term should be used.

In a quantitative study, the data for each image frame would be corrected according to the appropriate values for t and Δt and for the half-life $T_{1/2}$. For computational simplicity and efficiency, various approximations can be used when the parameter x in Equation 4-16 is small. For example, the following approximation is accurate to within 1% when $x < 0.25$:

$$
\mathrm{DF}_{\mathrm{eff}}\ (t, \Delta t) \approx \mathrm{DF}(t) \times [1 - (x/2)] \quad (4\text{-}17)
$$

where x again is defined as in Equation 4-16.

Another approach is to use the standard DF (see Equation 4-10) for the midpoint of the frame:

$$
\mathrm{DF}_{\mathrm{eff}}\ (t, \Delta t) \approx \mathrm{DF}\ [t + (\Delta t/2)] \quad (4\text{-}18)
$$

This approximation is accurate to within 1% for $x < 0.5$.

Yet another possibility is to use the average of the standard decay factors for the beginning and end of the frame:

$$
\mathrm{DF}_{\mathrm{eff}}\ (t, \Delta t) \approx \frac{[\mathrm{DF}(t) + \mathrm{DF}(t + \Delta t)]}{2} \quad (4\text{-}19)
$$

This approximation is accurate to within 1% for $x < 0.35$.

EXAMPLE 4-5

What are the effective decay factor and decay correction factor for the counts recorded in an image frame starting 30 sec and ending 45 sec after injection in a study performed with $^{15}\mathrm{O}$? Compare the results obtained with Equation 4-15 and the approximation

given by Equation 4-17. Assume that the data are to be corrected to $t = 0$, the time of injection.

Answer

From Appendix C, the half-life of ^{15}O is 122 sec. The decay factor at the beginning of the image frame, $t = 30$ sec, is

$$DF(30 \text{ sec}) = e^{-\ln 2 \times 30 \text{ sec}/122 \text{ sec}}$$

$$\approx e^{-0.170}$$

$$\approx 0.843$$

The duration of the image frame is $\Delta t = 15$ sec. The parameter x (Equation 4-16) is given by

$$x = \ln 2 \times (\Delta t/T_{1/2})$$

$$= \ln 2 \times (15 \text{ sec}/122 \text{ sec})$$

$$\approx 0.0852$$

Thus, decay during the image frame is given by

$$(1 - e^{-x})/x = (1 - e^{-0.0852})/0.0852$$

$$\approx 0.0817/0.0852$$

$$\approx 0.959$$

Taking the product of the two decay factors gives

$$DF_{\text{eff}} \approx 0.843 \times 0.959$$

$$\approx 0.808$$

The decay correction factor to apply to the counts recorded in this frame is

$$CF \approx 1/0.808$$

$$\approx 1.237$$

Using the approximation given by Equation 4-17 yields

$$DF_{\text{eff}} \approx 0.843 \times [1 - (0.0852/2)]$$

$$\approx 0.843 \times 0.957$$

$$\approx 0.807$$

which differs from the exact result obtained with Equation 4-15 by only approximately 0.1%.

E. SPECIFIC ACTIVITY

....

A radioactive sample may contain stable isotopes of the element represented by the radionuclide of interest. For example, a given ^{131}I sample may also contain the stable isotope

^{127}I. When stable isotopes of the radionuclide of interest are present in the sample, they are called *carrier*, and the sample is said to be *with carrier*. A sample that does not contain stable isotopes of the element represented by the radionuclide is called *carrier-free*.* Radionuclides may be produced carrier-free or with carrier, depending on the production method (see Chapter 5).

The ratio of radioisotope activity to total mass of the element present is called the *specific activity* of the sample. Specific activity has units of becquerels per gram, megabecquerels per gram, and so forth. The highest possible specific activity of a radionuclide is its *carrier-free specific activity* (CFSA). This value can be calculated in a straightforward manner from the basic properties of the radionuclide.

Suppose a carrier-free sample contains 1 g of a radionuclide ^{A}X, having a half-life $T_{1/2}$ (sec). The atomic weight of the radionuclide is approximately equal to A, its mass number (see Chapter 2, Section D.2). A sample containing A g of the radionuclide has approximately 6.023×10^{23} atoms (Avogadro's number); therefore a 1-g sample has $N \approx 6.023 \times 10^{23}/A$ atoms. The decay rate of the sample is $\Delta N/\Delta t$ (dps) $= \lambda N = 0.693N/T_{1/2}$. Therefore the activity per gram is:

$$A(\text{Bq/g}) \approx \ln 2 \times 6.023 \times 10^{23}/(A \times T_{1/2}) \quad (4\text{-}20)$$

Because the sample contains 1 g of the radioisotope, this is also its specific activity in becquerels per gram. When the equation is normalized for the half-life in *days* (1 day = 86,400 sec), the result is

$$\text{CFSA (Bq/g)} \approx 4.8 \times 10^{18}/(A \times T_{1/2}) \quad (4\text{-}21)$$

where $T_{1/2}$ is given in days. With appropriate normalization, Equation 4-21 also applies for specific activity in kBq/mg, GBq/g, and so on.

In radiochemistry applications, specific activities sometimes are specified in becquerels per mole of labeled compound. Because 1 mole of compound contains A g of radionuclide, this quantity is

$$\text{CFSA (Bq/mole)} = \text{CFSA (Bq/g)} \times A \text{ (g/mole)}$$

$$\approx 4.8 \times 10^{18}/T_{1/2} \quad (4\text{-}22)$$

where $T_{1/2}$ again is in days.

---

*Because it is virtually impossible to prepare a sample with absolutely no other atoms of the radioactive element, the terminology *without carrier* sometimes is used as well.

In traditional units, the equations for CFSA are

$$\text{CFSA (Ci / g)} \approx 1.3 \times 10^8 / (A \times T_{1/2})$$
$$\text{CFSA (Ci / mole)} \approx 1.3 \times 10^8 / T_{1/2}$$

(4-23)

where $T_{1/2}$ again is in days.

EXAMPLE 4-6

What are the CFSAs of ^{131}I and ^{99m}Tc?

Answer
For ^{131}I, A = 131 and $T_{1/2}$ = 8 days. Using Equation 4-21,

$$\text{CFSA} (^{131}\text{I}) \approx \frac{(4.8 \times 10^{18})}{(1.31 \times 10^2 \times 8)}$$
$$\approx 4.6 \times 10^{15} \text{ Bq / g}$$

For ^{99m}Tc, A = 99 and $T_{1/2}$ = 6 hours = 0.25 days. Thus,

$$\text{CFSA} (^{99m}\text{Tc}) \approx \frac{(4.8 \times 10^{18})}{(0.99 \times 10^2 \times 0.25)}$$
$$\approx 1.94 \times 10^{17} \text{ Bq / g}$$

In traditional units (Equation 4-23), the answers are

$$\text{CFSA} (^{131}\text{I}) \approx \frac{(1.3 \times 10^8)}{(1.31 \times 10^2 \times 8)}$$
$$\approx 1.24 \times 10^5 \text{ Ci / g}$$
$$\text{CFSA} (^{99m}\text{Tc}) \approx \frac{(1.3 \times 10^8)}{(0.99 \times 10^2 \times 0.25)}$$
$$\approx 5.3 \times 10^6 \text{ Ci / g}$$

As shown by Example 4-6, CFSAs for radionuclides having half-lives of hours, days, or even weeks are very high. Most of the radionuclides used in nuclear medicine are in this category.

In most instances, a high specific activity is desirable because then a moderate amount of activity contains only a very small mass of the element represented by the radioisotope and can be administered to a patient without causing a pharmacologic response to that element. This is an essential requirement of a "tracer study." For example, a capsule containing 0.4 MBq (~10 μCi) of carrier-free ^{131}I contains only approximately 10^{-10} g of elemental iodine (mass = activity/specific activity), which is well below the amount necessary to cause any "iodine reaction." Even radio-isotopes of highly toxic elements, such as arsenic, have been given to patients in a carrier-free state. It is not possible to obtain carrier-free ^{99m}Tc because it cannot be separated from its daughter product, ^{99}Tc, a very long-lived and essentially stable isotope of technetium. Nevertheless, the mass of technetium in most ^{99m}Tc preparations is very small and has no physiologic effect when administered to a patient.

Not all production methods result in carrier-free radionuclides. Also, in some cases carrier may be added to promote certain chemical reactions in radiochemistry procedures. When a preparation is supplied with carrier, usually the packaging material indicates specific activity. If the radioactivity exists as a label attached to some complex molecule, such as a protein molecule, the specific activity may be expressed in terms of the activity per unit mass of labeled substance, such as MBq/g of protein. Methods of calculating the specific activities of radionuclides produced in a non–carrier-free state are discussed in Chapter 5.

On rare occasions, radioactive preparations that are not carrier-free or that are attached as labels to complex molecules may present problems if the carrier or labeled molecule is toxic or has undesired pharmacologic effects. Two examples in the past were reactor-produced ^{42}K in K$^+$ solution (intravenous K$^+$ injections may cause cardiac arrhythmia) and ^{131}I-labeled serum albumin (serum albumin could cause undesirably high protein levels when injected into intrathecal spaces for cerebrospinal fluid studies). In situations such as these, the amount of material that can be administered safely to a patient may be limited by the amount of carrier or unlabeled molecule present rather than by the amount of radioactivity and associated radiation hazards.

F. DECAY OF A MIXED RADIONUCLIDE SAMPLE

The equations and methods presented in Sections B and C apply only to samples containing a single radionuclide species. When a sample contains a mixture of *unrelated* species (i.e., no parent-daughter relationships), the total activity A_t is just the sum of the individual activities of the various species:

$$A_t(t) = A_1(0)e^{-0.693t/T_{1/2,1}} + A_2(0)e^{-0.693t/T_{1/2,2}} + \cdots$$

(4-24)
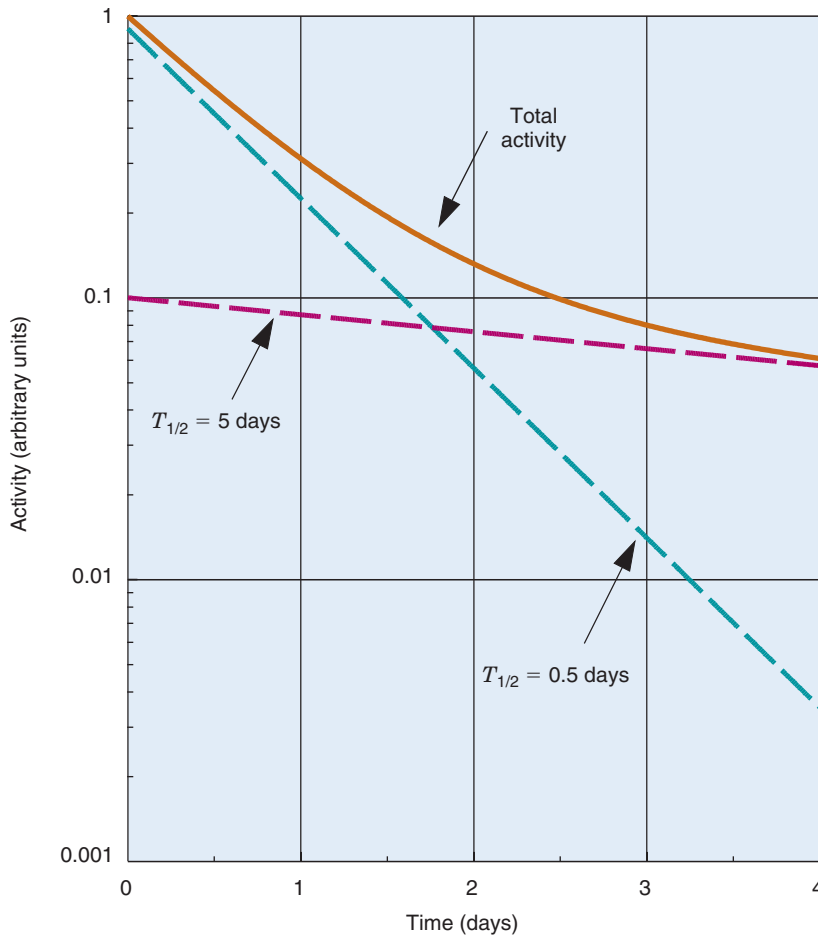
FIGURE 4-5 Activity versus time for a mixed sample of two unrelated radionuclides. The sample contains initially (at $t = 0$) 0.9 units of activity with a half-life of 0.5 days and 0.1 units of activity with a half-life of 5 days.

where $A_1(0)$ is the initial activity of the first species and $T_{1/2,1}$ is its half-life, and so forth.

Figure 4-5 shows total activity versus time for a sample containing two unrelated radionuclides. A characteristic of such a curve is that it *always* eventually follows the slope of the curve for the radionuclide having the longest half-life. Once the final slope has been established, it can be extrapolated as a straight line on a semilogarithmic graph back to time zero. This curve can then be subtracted from the total curve to give the net curve for the other radionuclides present. If more than two radionuclide species are present, the "curve-stripping" operation can be repeated for the next-longest-lived species and so forth.

Curve stripping can be used to determine the relative amounts of various radionuclides present in a mixed sample and their half-lives. It is especially useful for detecting and quantifying long-lived contaminants in radioactive preparations (e.g., [99]Mo in ^{99m}Tc).

G. PARENT-DAUGHTER DECAY

1. The Bateman Equations

A more complicated situation occurs when a sample contains radionuclides having parent-daughter relationships (Fig. 4-6). The
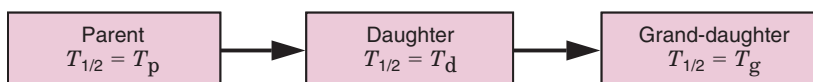
FIGURE 4-6 Schematic representation of series decay. Activities of the parent (p), daughter (d), and grand-daughter (g) are described by the Bateman equations.

equation for the activity of the parent is simply that for a single radionuclide species (see Equation 4-7); however, the equation for the activity of a daughter is complicated by the fact that the daughter product is being formed (by decay of the parent) at the same time it is decaying. The equation is

$$A_d(t) = \left\{ \left[A_p(0)\frac{\lambda_d}{\lambda_d - \lambda_p} \times \left(e^{-\lambda_p t} - e^{-\lambda_d t}\right)\right] \times \text{B.R.} \right\} + A_d(0)e^{-\lambda_d t}$$

$$(4\text{-}25)$$

where $A_p(t)$ and $A_d(t)$ are the activities of the parent and daughter radionuclides at time t, respectively, λ_p and λ_d are their respective decay constants, and B.R. is the branching ratio for decay to the daughter product of interest when more than one decay channel is available (see Equation 4-3).* The second

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*The differential equations from which Equation 4-25 is derived are

$$dN_p/dt = -\lambda_p N_p \qquad (4\text{-}25a)$$

$$dN_d/dt = -\lambda_d N_d + \lambda_p N_p \qquad (4\text{-}25b)$$

These equations provide

$$N_d(t) = N_p(0)\frac{\lambda_p}{\lambda_d - \lambda_p} \times (e^{-\lambda_p t} - e^{-\lambda_d t}) + N_d(0)e^{-\lambda_d t} \qquad (4\text{-}25c)$$

Multiplying Equation 4-25c by λ_d and substituting $A_d = \lambda_d N_d$, $A_p = \lambda_p N_p$, one obtains Equation 4-25.

term in Equation 4-25, $A_d(0)e^{-\lambda_d t}$, is just the residual daughter-product activity remaining from any that might have been present at time $t = 0$. In the rest of this discussion, it is assumed that $A_d(0) = 0$, and only the first term in Equation 4-25 is considered.

Equation 4-25 is the *Bateman equation* for a parent-daughter mixture. Bateman equations for sequences of three or more radionuclides in a sequential decay scheme are found in other texts.[1] Equation 4-25 is analyzed for three general situations.†
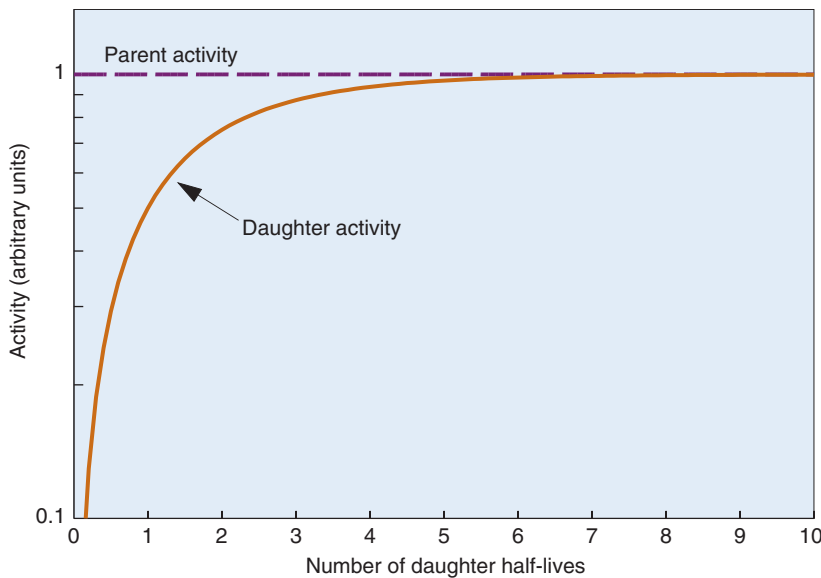
2. Secular Equilibrium

The first situation applies when the half-life of the parent, T_p, is so long that the decrease of parent activity is negligible during the course of the observation period. An example is ^{226}Ra ($T_p = 1620$ yr) → ^{222}Rn ($T_d = 4.8$ days). In this case, $\lambda_p \approx 0$; thus Equation 4-25 can be written

$$A_d(t) \approx A_p(0)(1 - e^{-\lambda_d t}) \times \text{B.R.} \qquad (4\text{-}26)$$

Figure 4-7 illustrates the buildup of daughter product activity versus time for B.R. = 1. After one daughter-product half-life, $e^{-\lambda_d t} = 1/2$

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†A fourth (but unlikely) situation occurs when $\lambda_p = \lambda_d = \lambda$, that is when parent and daughter have the same half-life. In this case, it can be shown that Equation 4-25 reduces to

$$A_d(t) = A_p(0)te^{-\lambda t} + A_d(0)e^{-\lambda t} \qquad (4\text{-}25d)$$

FIGURE 4-7   Buildup of daughter activity when $T_d \ll T_p \approx \infty$, branching ratio = 1. Eventually, secular equilibrium is achieved.

and $A_d \approx (1/2)A_p$. After two half-lives, $A_d \approx (3/4)A_p$, and so forth. After a "very long" time, $(\sim 5 \times T_d)$, $e^{-\lambda_d t} \approx 0$, and the activity of the daughter equals that of the parent. When this occurs ($A_d \approx A_p \times$ B.R.), the parent and daughter are said to be in *secular equilibrium.*

3. Transient Equilibrium

The second situation occurs when the parent half-life is longer than the daughter half-life but is not "infinite." An example of this case is ^{99}Mo ($T_{1/2} = 66$ hr) $\rightarrow$ ^{99m}Tc ($T_{1/2} = 6$ hr). When there is a significant decrease in parent activity during the course of the observation period, one can no longer assume $\lambda_p \approx 0$, and Equation 4-25 cannot be simplified. Figure 4-8 shows the buildup and decay of daughter-product activity for a hypothetical parent-daughter pair with $T_p = 10 T_d$ and B.R. = 1. The daughter-product activity increases and eventually exceeds that of the parent, reaches a maximum value, and then decreases and follows the decay of the parent. When this stage of "parallel" decay rates has been reached—that is, parent and daughter activities are decreasing but the *ratio* of parent-to-daughter activities are constant—the parent and daughter are said to be in *transient equilibrium*. The ratio of daughter-to-parent activity in transient equilibrium is

$$A_d/A_p = [T_p/(T_p - T_d)] \times \text{B.R.} \quad (4\text{-}27)$$

The time at which maximum daughter activity is available is determined using the methods of calculus* with the result

$$t_{max} = [1.44 T_p T_d/(T_p - T_d)]\ln(T_p/T_d) \quad (4\text{-}28)$$

where T_p and T_d are the half-lives of the parent and daughter, respectively.

Figure 4-8 is similar to that for ^{99}Mo ($T_p = 66$ hr) $\rightarrow$ ^{99m}Tc ($T_d = 6$ hr); however, the time-activity curve for ^{99m}Tc is somewhat lower because only a fraction (B.R. = 0.876) of the parent ^{99}Mo atoms decay to ^{99m}Tc (see Fig. 5-7). The remainder bypass the ^{99m}Tc metastable state and decay directly to the ground state of ^{99}Tc. Thus the ^{99m}Tc activity is given by Equation 4-25 multiplied by 0.876 and the ratio of ^{99m}Tc/^{99}Mo activity in transient equilibrium by Equation 4-27 multiplied by the same factor; however, t_{max} remains as given by Equation 4-28.
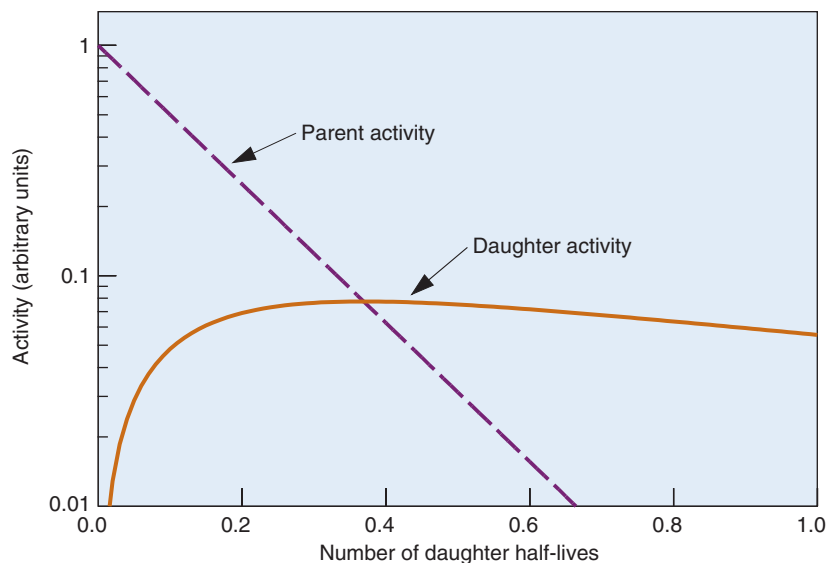
4. No Equilibrium

When the daughter half-life is longer than the parent half-life, there is no equilibrium between them. An example of this combination is ^{131m}Te ($T_{1/2} = 30$ hr) $\rightarrow$ ^{131}I ($T_{1/2} = 8$ days). Figure 4-9 shows the buildup and

*Set $dA_d/dt = 0$ and solve for t_{max}.
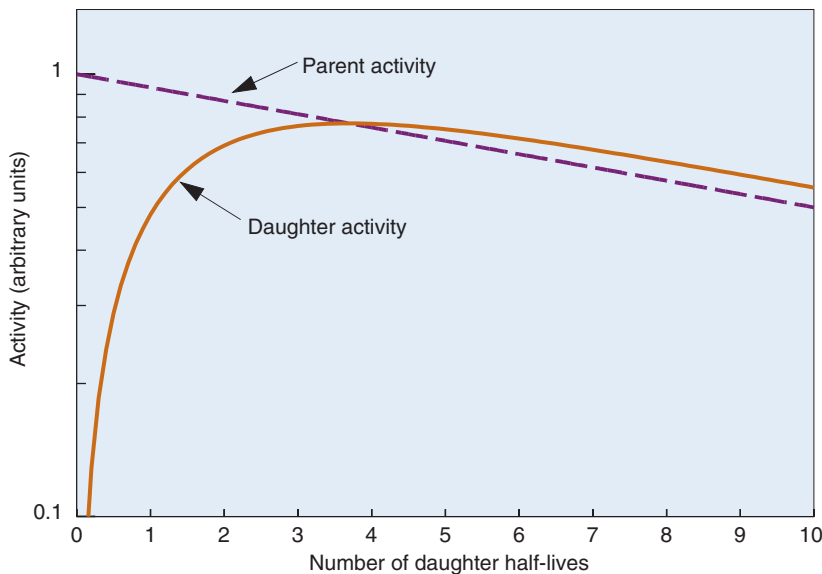
FIGURE 4-8 Buildup and decay of activity for $T_p = 10 T_d$, branching ratio = 1. Eventually, transient equilibrium is achieved when the parent and daughter decay curves are parallel.

FIGURE 4-9   Buildup and decay of activity for $T_p = 0.1\ T_d$, branching ratio = 1. There is no equilibrium relationship established between the parent and daughter decay curves.

decay of the daughter product activity for a hypothetical parent-daughter pair with $T_p = 0.1T_d$. It increases, reaches a maximum (Equation 4-28 still applies for t_{max}), and then decreases. Eventually, when the parent activity is essentially zero, the remaining daughter activity decays with its own characteristic half-life.

REFERENCE

1. Evans RD: *The Atomic Nucleus*, New York, 1972, McGraw-Hill, pp 477-499.