1

Radioactive Decay and the Origin of Gamma and X-Radiation

1.1 Introduction

In this chapter, I intend to show how a basic understanding of simple decay schemes, and of the role gamma radiation plays in these, can help in identifying radioactive nuclides and in correctly measuring quantities of such nuclides. In doing so, I need to introduce some elementary concepts of nuclear stability and radioactive decay. X-radiation can be detected by using the same or similar equipment, and I will also discuss the origin of X-rays in decay processes and the light that this knowledge sheds on characterization procedures.

I will show how the Karlsruhe Chart of the Nuclides can be of help in predicting or confirming the identity of radionuclides, being useful both for the modest amount of nuclear data it contains and for the ease with which generic information as to the type of nuclide expected can be seen.

First, I will briefly look at the nucleus and nuclear stability. I will consider a nucleus simply as an assembly of uncharged neutrons and positively charged protons; both of these are called **nucleons**.

Number of neutrons $= N$

Number of protons $= Z$

Z is the **atomic number** and defines the element. In the neutral atom, Z will also be the number of extranuclear electrons in their atomic orbitals. An element has a fixed Z, but in general will be a mixture of atoms with different masses, depending on how many neutrons are present in each nucleus. The total number of nucleons is called the **mass number**.

Mass number $= N + Z = A$

A, N and Z are all integers by definition. In practice, a neutron has a very similar mass to a proton and so there is a real physical justification for this usage. In general, an assembly of nucleons should be referred to as a **nuclide**. Conventionally, a nuclide of atomic number Z and mass number A is specified as ${}^{A}_{Z}\mathrm{Sy}$, where Sy is the chemical symbol of the element. Thus, ${}^{58}_{27}\mathrm{Co}$ is a nuclide with 27 protons and 31 neutrons. Because the chemical symbol uniquely identifies the element, unless there is a particular reason for including it, the atomic number as subscript is usually omitted – as in ${}^{58}\mathrm{Co}$. This format leaves room for chemical information to follow; for example, ${}^{58}\mathrm{Co}^{2+}$. As it happens, this particular nuclide is radioactive and could, in order to impart that extra item of knowledge, be referred to as a **radionuclide**. Unfortunately, in the world outside of physics and radiochemistry, the word **isotope** has become synonymous with radionuclide – something dangerous and unpleasant. In fact, isotopes are simply atoms of the same element (i.e. same Z, different N) – radioactive or not. Thus, ${}^{58}_{27}\mathrm{Co}$, ${}^{59}_{27}\mathrm{Co}$ and ${}^{60}_{27}\mathrm{Co}$ are isotopes of cobalt. Here, 27 is the atomic number, and 58, 59 and 60 are **mass numbers** equal to the total number of nucleons. ${}^{59}\mathrm{Co}$ is stable; it is, in fact, the only stable isotope of cobalt.

Returning to nomenclature, ${}^{58}\mathrm{Co}$ and ${}^{60}\mathrm{Co}$ are **radioisotopes**, as they are unstable and undergo radioactive decay. It would be incorrect to say, 'the radioisotopes ${}^{60}\mathrm{Co}$ and ${}^{239}\mathrm{Pu}$…' because two different elements are being discussed; the correct expression would be 'the radionuclides ${}^{60}\mathrm{Co}$ and ${}^{239}\mathrm{Pu}$…'.

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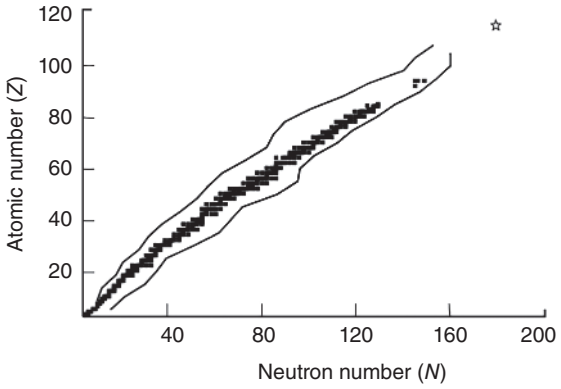


Figure 1.1 A Segrè chart. The symbols mark all known stable nuclides as a function of Z and N. At high Z, the long half-lived Th and U nuclides are shown. The outer envelope encloses known radioactive species. The star marks the position of the heaviest known nuclide, ^{294}Og.

If all stable nuclides are plotted as a function of Z (y-axis) and N (x-axis), then Figure 1.1 will result. This is a **Segrè chart**. The star marks the position of the largest nuclide known to date, $^{294}_{118}$Og, oganesson with 118 protons. Named in 2016, it is one of the only two elements named after a person who was alive at the time of naming; the other being seaborgium and the only element whose eponym was alive at the time of writing (2023).

The **Karlsruhe Chart of the Nuclides** has this same basic structure but with the addition of all known radioactive nuclides. The heaviest stable element is bismuth ($Z = 83$, $N = 126$). The figure also shows the location of some high-Z unstable nuclides – the major thorium ($Z = 90$) and uranium ($Z = 92$) nuclides. Theory has predicted that there could be stable nuclides, as yet unknown, called *superheavy nuclides* on an **island of stability** at about $Z = 114$, $N = 184$, well above the current known range.

Radioactive decay is a spontaneous change within the nucleus of an atom that results in the emission of particles. The modes of radioactive decay are principally alpha and beta decays, with spontaneous fission as one of a small number of rarer processes. Radioactive decay is driven by mass change – the mass of the product or products is smaller than the mass of the original nuclide. Decay is always exoergic; the small mass change appears as energy in an amount determined by the following equation introduced by Einstein:

$$\Delta E = \Delta m \times c^2$$

where the energy difference is in joules, the mass in kilograms and the speed of light in ms^{-1}.

The unit of energy we use in gamma spectrometry is the electron-volt (eV), where $1\,eV = 1.602\,177 \times 10^{-19}$ J.[1] Hence, $1\,eV = 1.782\,663 \times 10^{-36}$ kg or $1.073\,533 \times 10^{-9}$ u ('u' is the unit of atomic mass (amu), defined as 1/12th of the mass of ^{12}C). Energies in the gamma radiation range are conveniently in keV.

Gamma-ray emission is not, strictly speaking, a decay process; it is a de-excitation of the nucleus. I will now explain each of these decay modes and show, in particular, how gamma emission frequently appears as a by-product of alpha or beta decay, being one way in which residual excitation energy is dissipated.

1.2 Beta Decay

Figure 1.2 shows a three-dimensional version of the low-mass end of the Segrè chart with energy/mass plotted on the third axis, shown vertically here. We can think of the stable nuclides as occupying the bottom of a nuclear-stability valley that runs from hydrogen to bismuth. The stability can be explained in terms of particular relationships between Z and N. Nuclides outside this valley bottom are unstable and can be imagined as sitting on the sides of the valley at heights that reflect their relative nuclear masses or energies.

1 Values given are rounded from those recommended by the UK National Physical Laboratory in *Fundamental Physical Constants and Energy Conversion Factors* (1991).

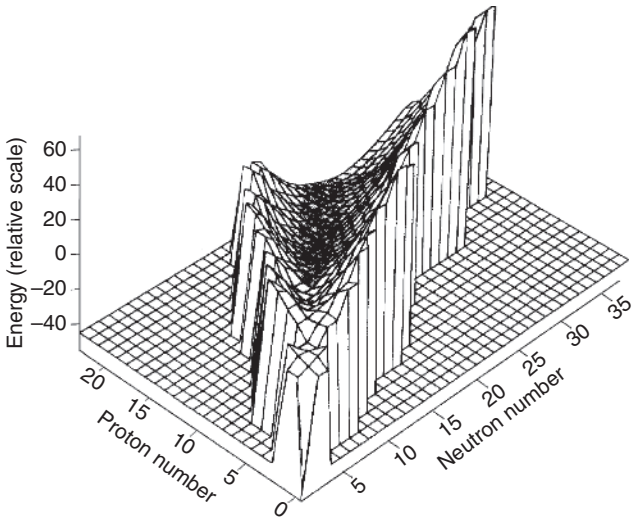
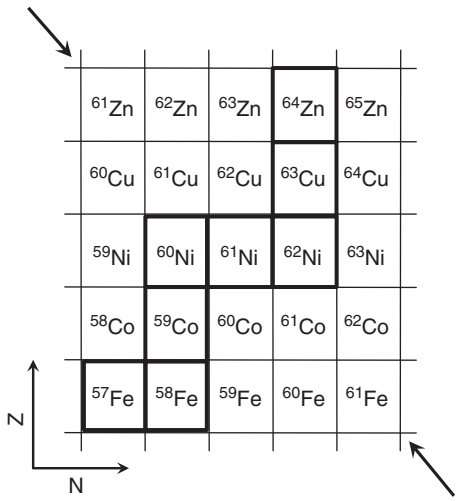


Figure 1.2 The beta stability valley at low Z (adapted from a figure published by *New Scientist*, and reproduced with permission).

Figure 1.3 Part of the Chart of the Nuclides. Heavy boxes indicate the stable nuclides.



The dominant form of radioactive decay is movement down the hillside directly to the valley bottom. This is **beta decay**. It corresponds to transitions along an **isobar** or line of constant A. What is happening is that neutrons are changing to protons (β^- decay), or, on the opposite side of the valley, protons are changing to neutrons (β^+ decay or electron capture). Figure 1.3 is part of the (Karlsruhe) Nuclide Chart.

If we consider the isobar $A = 61$, ^{61}Ni is stable, and beta decay can take place along a diagonal (in this format) from either side. ^{61}Ni has the smallest mass in this sequence, and the driving force is the mass difference; this appears as energy released. These energies are shown in Figure 1.4. There are theoretical grounds, based on the liquid drop model of the nucleus, for thinking that these points fall on a parabola.

1.2.1 β^- or Negatron Decay

The decay of ^{60}Co is an example of β^- or **negatron** decay (negatron = negatively charged beta particle). All nuclides unstable to β^- decay are on the neutron-rich side of stability. (On the Karlsruhe Chart, these are coloured

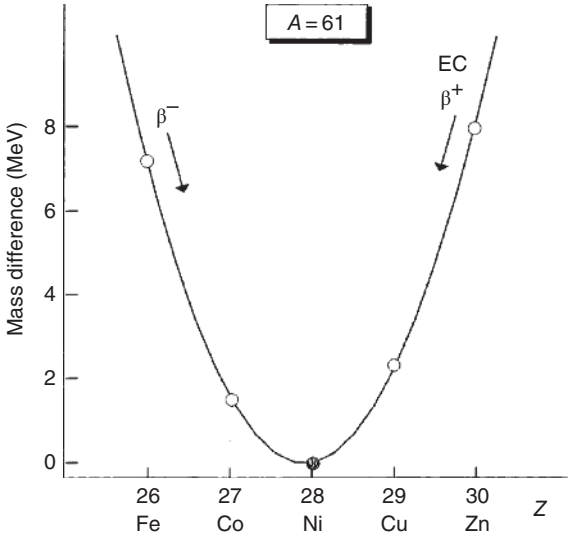


Figure 1.4 The energy parabola for the isobar $A = 61$. ^{61}Ni is stable, while other nuclides are beta-active (EC, electron capture).

blue.) The decay process addresses that instability. An example of β^- decay is as follows:

$$^{60}\text{Co} \rightarrow {}^{60}\text{Ni} + \beta^- + \bar{\nu}$$

A **beta particle**, β^-, is an electron; in all respects, it is identical to any other electron. Following on from Section 1.1, the sum of the masses of the ^{60}Ni plus the mass of the β^-, and $\bar{\nu}$, the anti-neutrino, are less than the mass of ^{60}Co. That mass difference drives the decay and appears as the energy of the decay products. What happens during the decay process is that a neutron is converted to a proton within the nucleus. In that way, the atomic number increases by one, and the nuclide drops down the side of the valley to a more stable condition. A fact not often realized is that the neutron itself is radioactive when it is not bound within a nucleus. A free neutron has a half-life of only 10.2 m and decays by beta emission:

$$\text{n} \rightarrow \text{p}^+ + \beta^- + \bar{\nu}$$

That process is essentially the conversion process happening within the nucleus.

The decay energy is shared between the particles in inverse ratio to their masses in order to conserve momentum. The mass of ^{60}Ni is very large compared with the mass of the beta particle and neutrino and, from a gamma spectrometry perspective, takes a very small, insignificant portion of the decay energy. The beta particle and the anti-neutrino share almost the whole of the decay energy in variable proportions; each takes from 0% to 100% in a statistically determined fashion. For that reason, beta particles are not mono-energetic, as one might expect from the decay scheme, and their energy is usually specified as $E_{\beta\text{max}}$. The term 'beta particle' is reserved for an electron that has been emitted during a nuclear decay process. This distinguishes it from electrons emitted as a result of other processes, which will usually have defined energies. The anti-neutrino need not concern us as it is detectable only in elaborate experiments. Anti-neutrinos (and neutrinos from β^+ decay) are theoretically crucial in maintaining the universality of the conservation laws of energy and angular momentum.

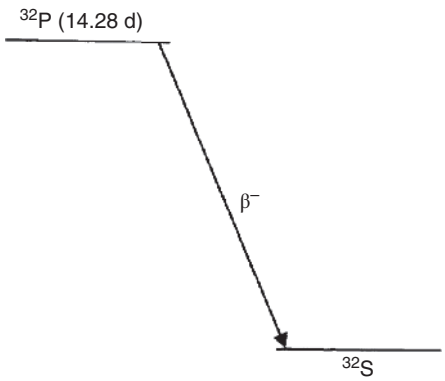
The lowest energy state of each nuclide is called the **ground state**, and it would be unusual for a transition to be made directly from one ground state to the next – unusual, but unfortunately far from unknown. There are a number of technologically important pure beta emitters, which are either widely used as radioactive tracers (^{3}H, ^{14}C, ^{35}S) or have significant yields in fission (^{90}Sr/^{90}Y, ^{99}Tc, ^{147}Pm). Table 1.1 lists the most common beta emitters along with their half-lives. The decay scheme of all of the nuclides in Table 1.1 will be of the form shown in Figure 1.5.

Table 1.1 Some pure beta emitters[a].

Nuclide	Half-life[b)c)]	Maximum beta energy (keV)
^{3}H	12.312 (25) a	19
^{14}C	5700 (30) a	156
^{32}P	14.284 (36) d	1711
^{35}S	87.32 (16) d	167
^{36}Cl	$3.01\ (2) \times 10^{5}$ a	1142
^{45}Ca	162.61 (9) d	257
^{63}Ni	98.7 (24) a	66
^{90}Sr	28.80 (7) a	546
^{90}Y	2.6684 (13) d	2282
^{99}Tc	$2.111\ (12) \times 10^{5}$ a[b)]	294
^{147}Pm	2.6234 (2) a[b)]	225
^{204}Tl	3.788 (15) a	763

a) Data taken from *DDEP* (1986).
b) Data taken from *Table of Isotopes* (1978, 1998).
c) Figures in parentheses represent the 1σ uncertainties on the last digit or digits.

Figure 1.5 The decay scheme of a pure beta emitter, ^{32}P.



You should be aware that because the 'year' has alternative definitions, it is now conventional to use the 'day' as the unit for quoting long half-lives. You should also be aware that the international abbreviation for the year is 'a' – after the French word for year – 'an'.

The difficulty for gamma spectrometrists is that no gamma radiation is emitted by these radionuclides and thus they cannot be measured by the techniques described in this text. To determine pure beta emitters in a mixture of radionuclides, a degree of chemical separation is required, followed by measurement of the beta radiation, perhaps by liquid scintillation or by using a gas-filled detector.

However, many beta transitions do not go to the ground state of the daughter nucleus, but to an excited state. This behaviour can be seen superimposed on the isobaric energy parabola in Figure 1.6. Excited states are shown for both radioactive (Ag, Cd, In, Sb, Te) and stable (Sn) isobaric nuclides, and it should be noted that these states are approached through the preceding or parent nuclide.

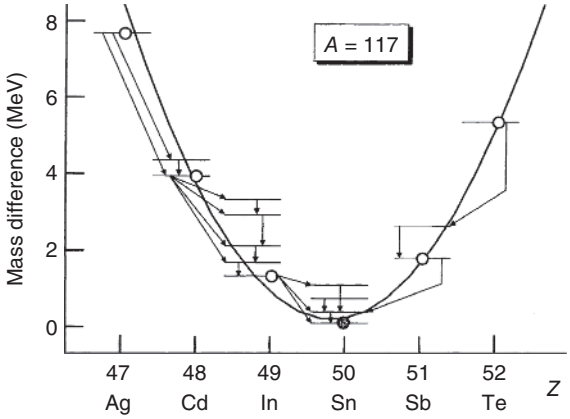


Figure 1.6 The isobar $A = 117$ with individual decay schemes superimposed. ^{117}Sn is stable.

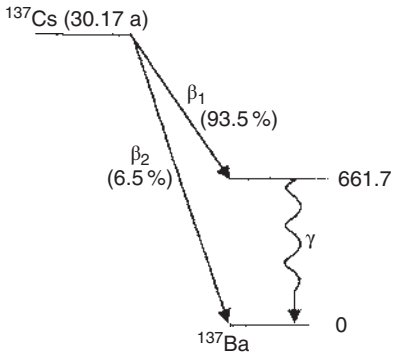


Figure 1.7 The decay scheme of ^{137}Cs.

The **decay scheme** for a single beta-emitting radionuclide is part of this energy parabola with just the two components of parent and daughter. Figure 1.7 shows the simple case of ^{137}Cs. Here, some beta decays (6.5% of the total) go directly to the ground state of ^{137}Ba; most (93.5%) go to an excited nuclear state of ^{137}Ba.

The gamma radiation is released as that excited state de-excites and drops to the ground state. Note that the energy released, 661.7 keV, is actually a property of ^{137}Ba, but is accessed from ^{137}Cs. It is conventionally regarded as 'the ^{137}Cs gamma' and listed in data tables as such.

However, when looking for data about *energy levels* in the nucleus, as opposed to gamma-ray energies, it would be necessary to look under the daughter, ^{137}Ba.

In this particular case, 661.7 keV is the *only* gamma-ray emitted in the decay process. More commonly, many gamma transitions are involved. This is seen in Figures 1.6 and 1.8, where, for ^{60}Co, the great majority of beta decays (those labelled β_1) go to the 2505.7 keV level, which falls to the ground state in two steps. Thus, two gamma-rays

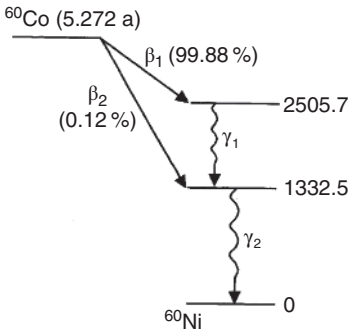


Figure 1.8 The decay scheme of ^{60}Co.

appear with their energies being the difference between the energies of the upper and lower levels:

$$\gamma_1 = (2505.7 - 1332.5) = 1173.2 \text{ keV}$$

$$\gamma_2 = (1332.5 - 0) = 1332.5 \text{ keV}$$

The two gammas are said to be **in cascade**, and if they appear at essentially the same time, that is, if the intermediate level (in ^{60}Ni at 1332.5 keV) does not delay emission of the second gamma, then they are also said to be **coincident**. This phenomenon of two gamma-rays appearing from the same atom at the same instant can have a significant influence on counting efficiency, as will be discussed in Chapter 8.

1.2.2 β^+ or Positron Decay

Just as β^- active nuclides are neutron rich, nuclides unstable to β^+ decay are neutron deficient. (The red nuclides on the Karlsruhe Chart.) The purpose of positron decay, again driven by mass difference, is to convert a proton into a neutron. Again, the effect is to slide down the energy parabola in Figure 1.4, this time on the neutron-deficient side, towards stability, resulting in an atom of a lower atomic number than the parent. An example is as follows:

$$^{64}_{29}\text{Cu} \rightarrow {}^{64}_{28}\text{Ni} + \beta^+ + \nu \text{ (neutrino)}$$

During this decay a positron, a positively charged electron (anti-electron), is emitted, and conservation issues are met by the appearance of a neutrino. Beta decay processes involve the transformation of a neutron into a proton or vice versa. The mechanism is related to a change in the underlying quark structure of the nucleon mediated by the weak force. Positron emission is only possible if there is a sufficiently large energy difference between the consecutive isobaric nuclides. This imposes a critical threshold for this decay mode, which requires the mass difference between the parent nuclide and the sum of the daughter mass, electron and the positron to be in excess of 1022 keV. As with negatrons, there is a continuous energy spectrum ranging up to a maximum value and emission of complementary neutrinos.

The positron has a short life; it is rapidly slowed in matter until it reaches a very low, close to zero, kinetic energy. Positrons are anti-particles to electrons, and the slowed positron will inevitably find itself near an electron. The couple may exist for a short time as **positronium** – then the process of **annihilation** occurs. Both the positron and electron disappear, and two photons are produced, each with energy equal to the electron mass, 511.00 keV (Figure 1.9). These photons are called **annihilation radiation** and the annihilation peak is a common feature in gamma spectra, which is much enhanced when β^+ nuclides are present. To conserve momentum, the two 511 keV photons will be emitted in exactly opposite directions. I will mention here, and treat the implications more fully later, that the annihilation peak in the spectrum will be considerably broader than a peak produced by a direct nuclear-generated gamma-ray of the same energy. This can help in distinguishing between the two. The reason for such broadening is due to the **Doppler effect**. At the point where the positron–electron interaction takes place, neither positron nor electron is likely to be at complete rest; the positron may have a small fraction of its initial kinetic energy and the electron – if we regard it as a particle circling the nucleus – because of its orbital momentum. Thus, there may well be a resultant net momentum of the particles at the moment of interaction, so

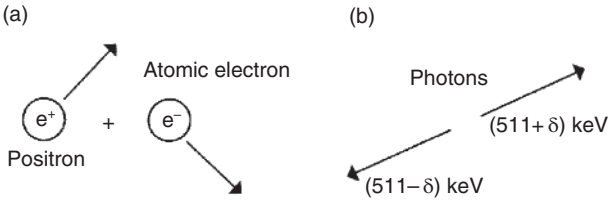


Figure 1.9 The annihilation process, showing how the resultant 511 keV photons could have a small energy shift: (a) possible momenta before interaction giving (b) differing photon energies after interaction.

that the conservation laws mean that one 511 keV photon will be slightly larger in energy and the other slightly smaller. This increases the statistical uncertainty and widens the peak. Note that the sum of the two will still be (in a centre of mass system) precisely 1022.00 keV.

1.2.3 Electron Capture (EC)

As described earlier, β^+ can only occur if more than 1022 keV of decay energy is available. For neutron-deficient nuclides close to stability where that energy is not available, an alternative means of decay is available. In this, the electron needed to convert the proton is captured by the nucleus from one of the extranuclear electron shells. The process is known as **electron capture** decay. As the K shell is closest to the nucleus (the wave functions of the nucleus and K shell have a greater degree of overlap than with more distant shells), the capture of a K electron is most likely to happen and indeed sometimes the process is called **K-capture**. The probability of capture from the less-strongly bound higher shells (L, M, etc.) increases as the decay energy decreases.

Loss of an electron from the K shell leaves a vacancy there (Figure 1.10). This is filled by an electron dropping in from a higher, less-tightly bound, shell. The energy released in this process often appears as an X-ray, in what is referred to as **fluorescence**. One X-ray may well be followed by others (of lower energy) as electrons cascade down from shell to shell towards greater stability.

Sometimes, the energy released in rearranging the electron structure does not appear as an X-ray. Instead, it is used to free an electron from the atom as a whole. This is the **Auger effect**, emitting **Auger electrons**. The probability of this alternative varies with Z: at higher Z, there will be more X-rays and fewer Auger electrons; it is said that the **fluorescence yield** is greater. Auger electrons are mono-energetic and usually of low energy, being emitted from an atomic orbital (L or M) where the electron-binding energies are smaller. There is a small probability of both Auger electrons and X-rays being emitted together in one decay; this is the radiative Auger effect. Note that whenever X-rays are emitted, they will be characteristic of the daughter, rather than the parent, as the rearrangement of the electron shells is occurring after the electron capture.

For neutron-deficient nuclides with a potential decay energy somewhat above the threshold value of 1022 keV, both positron decay and electron capture decay will occur, in a proportion statistically determined by the different decay energies of the two processes. Figure 1.11 shows the major components of the decay scheme of ^{22}Na, where both positron decay and electron capture are involved. We can deduce from this that the spectrum will show a gamma-ray at 1274.5 keV, an annihilation peak at 511.0 keV (from the β^+), and probably X-rays due to electron rearrangement after the EC.

1.2.4 Multiple Stable Isotopes

In Figures 1.4 and 1.6, I suggested that the ground states of the nuclides of isobaric chains lay on a parabola, and the decay involved moving down the sides of the parabola to the stable point at the bottom. The implication must be that there is only one stable nuclide per isobaric chain. Examination of the Karlsruhe Chart shows quite clearly that this is not true – there are many instances of two, or even three, stable nuclides on some isobars. More careful examination reveals that *what* is true is that every *odd*-isobar only has one stable nuclide. It is the even-numbered

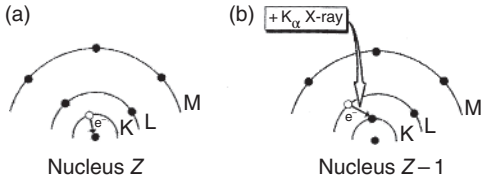


Figure 1.10 (a) Electron capture from the K shell, followed by (b) electron movement (X-ray emission) from L to K and then from M to L resulting in X-radiations.

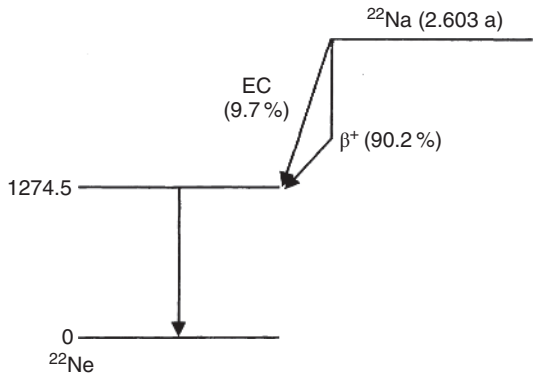


Figure 1.11 The decay scheme of ^{22}Na. Note the representation of positron emission, where 1022 keV is lost before emission of the β^+ particle.

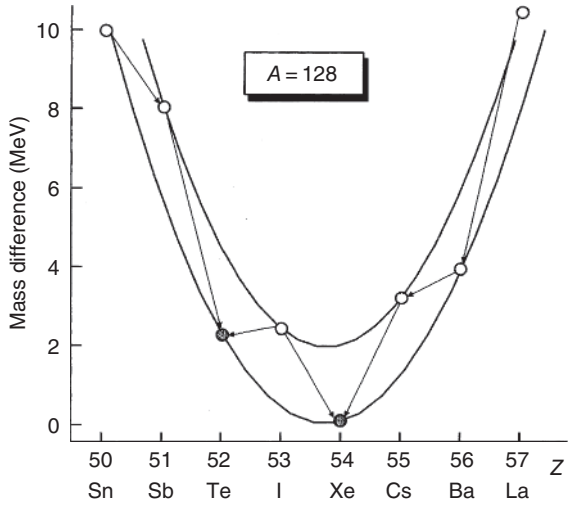


Figure 1.12 The two energy parabolas for the isobar $A = 128$. ^{128}Te and ^{128}Xe are stable.

isobars that are the problem. If a parabola can only have one bottom, the implication is that for even-isobars there must be more than one stability parabola. Indeed, that is so. In fact, there are two parabolas; one corresponding to even-Z/even-N (even–even) and the other to odd-Z/odd-N (odd–odd). Figure 1.12 shows this. The difference arises because pairing of nucleons gives a small increase in stability – a lowering of energy. In even–even nuclides, there are more paired nucleons than in odd–odd nuclides, and so the even–even parabola is lower in energy. As shown in Figure 1.12 for $A = 128$ isobaric chain, successive decays make the nucleus jump from odd–odd to even–even and back. There will be occasions, as here, where a nucleus finds itself above the ultimate lowest point of the even–even parabola, but below the neighbouring odd–odd points. It will, therefore, be stable. (It is the theoretical possibility that a nuclide such as ^{128}Te could decay to ^{128}Xe, which fuels the search for double beta decay, which I will refer to from time to time.) In all, depending upon the particular energy levels of neighbouring isobaric nuclides, there could be up to three stable nuclides per even-A isobaric chain.

In the case of $A = 128$, there are two stable nuclides, ^{128}Te and ^{128}Xe. ^{128}I has a choice of destination and, in 93.1% of cases, decays by β^- to ^{128}Xe and, in 6.98% cases, decays by EC to ^{128}Te. The dominance of the ^{128}Xe transition reflects the greater energy release, as indicated in Figure 1.12. This behaviour is quite common for even mass parabolas, and this choice of decay mode is available for well-known nuclides, such as ^{40}K and ^{152}Eu. Occasionally, if the decay energy is sufficient, a nuclide will decay sometimes by β^- and sometimes by EC and β^+.

1.3 Alpha Decay

An alpha particle is an He-4 nucleus, ${}^{4}_{2}He^{2+}$, and the emission of this particle is commonly the preferred mode of decay at high atomic numbers, $Z > 83$. In losing an alpha particle, the nucleus loses four units of mass and two units of charge:

$$Z \rightarrow Z - 2$$

$$A \rightarrow A - 4$$

Typical is the decay of the most common isotope of radium:

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

The product in this case is the most common isotope of radon, ^{222}Rn (usually just called 'radon' and which, incidentally, is responsible for the largest radiation dose from a single nuclide to the general population). A fixed quantity of energy, Q, equal to the difference in mass between the initial nuclide and final products, is released. This energy must be shared between the Rn and the He in a definite ratio because of the conservation of momentum. Thus, the alpha particle is mono-energetic and alpha spectrometry becomes possible. In contrast to beta decay, there are no neutrinos to take away a variable fraction of the energy.

In many cases, especially in the lower Z range of alpha decay, the emission of an alpha particle takes the nucleus directly to the ground state of the daughter, analogous to the 'pure-β' emission described earlier. However, with heavier nuclei, α decay can lead to excited states of the daughter. Figure 1.13, the decay scheme of ^{228}Th, shows gamma emission following alpha decay, but even here, it will be seen that most alpha transitions go directly to the ^{224}Ra ground state.

Calculation of the alpha decay energy reveals that even nuclides, such as ^{152}Eu and the stable ^{151}Eu, are unstable towards alpha decay. Alpha decay of ^{151}Eu would release 1.96 MeV of energy. The reason that this, and most other nuclides, do not decay by alpha emission is the presence of an energy barrier – it takes energy to prise an alpha particle out of the nucleus. Unless the nucleus is excited enough or the decay energy is sufficient to tunnel through the energy barrier, it will be stable to alpha emission. That does not preclude it from being unstable to beta decay; ^{151}Eu is stable, and ^{152}Eu is radioactive.

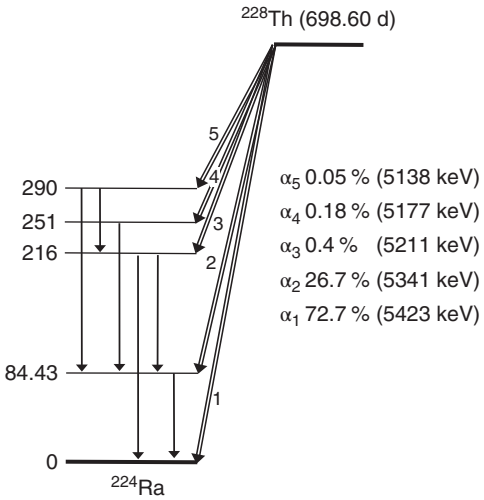


Figure 1.13 The decay scheme of ^{228}Th.

1.4 Spontaneous Fission (SF)

Spontaneous fission is a natural decay process in which a heavy nucleus spontaneously splits into two large fragments. An example is as follows:

$$^{252}_{98}\mathrm{Cf} \rightarrow {}^{140}_{54}\mathrm{Xe} + {}^{108}_{44}\mathrm{Ru} + 4\,{}^{1}_{0}\mathrm{n} + Q$$

The two product nuclides are only examples of what is produced; these are fission fragments or (when in their ground states) fission products. The range of products, the energies involved (Q) and the number and energies of neutrons emitted are all similar to those produced in more familiar neutron-induced fission of fissile or fissionable nuclides. ^{252}Cf is mentioned here as it is a commercially available nuclide, which is bought either as a source of fission fragments or as a source of neutrons.

Once more, the driving force for the process is the release of energy. Q is of the order of 200 MeV, a large quantity, showing that the fission products have a substantially smaller joint mass than the fissioning nucleus. This is because the binding energy per nucleon is significantly greater for nuclides in the middle of the periodic table than at the extremes. ^{108}Ru, for example, has a binding energy of about 8.55 MeV per nucleon, while the corresponding figure for ^{252}Cf is about 7.45 MeV per nucleon. Despite the emission of neutrons in fission, fission products are overwhelmingly likely to find themselves on the neutron-rich, β^- active side of the nuclear stability line. They will then undergo β^- decay along an isobar, as, for example, along the left-hand side of Figure 1.12, until a stable nucleus is reached. During this sequence, gamma emission is almost always involved, as described earlier. The distribution of fission product masses will be discussed in Section 1.9.

As with alpha decay, calculation of mass differences for notional fission outcomes suggests that even mid-range nuclides, in terms of mass, would be unstable for fission. Fission is prevented in all but very large nuclei by the **fission barrier**: the energy needed to deform the nucleus from a sphere to a situation where two nearly spherical fission product nuclei can split off.

1.5 Exotic Decay Modes

A number of uncommon decay modes, and sources of gamma-rays exist which are of little direct relevance to gamma spectrometrists, and I will content myself with just listing them: delayed neutron emission, delayed proton emission, proton emission, double beta decay (the simultaneous emission of two β^- particles), two proton decay and the emission of 'heavy ions' or 'clusters', such as ^{14}C and ^{24}Ne. There are also exotic decay processes involving sub-atomic particles which take place within our atmosphere, such as π-zero decay, that release high-energy gamma-rays. Some detail can be found in the more general texts in Further Reading section, such as the one by Ehmann and Vance (1991).

1.6 Gamma Emission

This is not a form of decay like alpha, beta or spontaneous fission, in that there is no change in the number or type of nucleons in the nucleus; there is no change in Z, N or A. The process is solely that of losing surplus excitation energy and, as I have shown, is usually a by-product of alpha or beta decay. First – what is a gamma-ray?

1.6.1 The Electromagnetic Spectrum

Gamma radiation is electromagnetic radiation, basically just like radio waves, microwaves and visible light. In the enormous range of energies in the electromagnetic spectrum, gamma-rays sit at the high-energy, short-wavelength end, as shown in Figure 1.14.

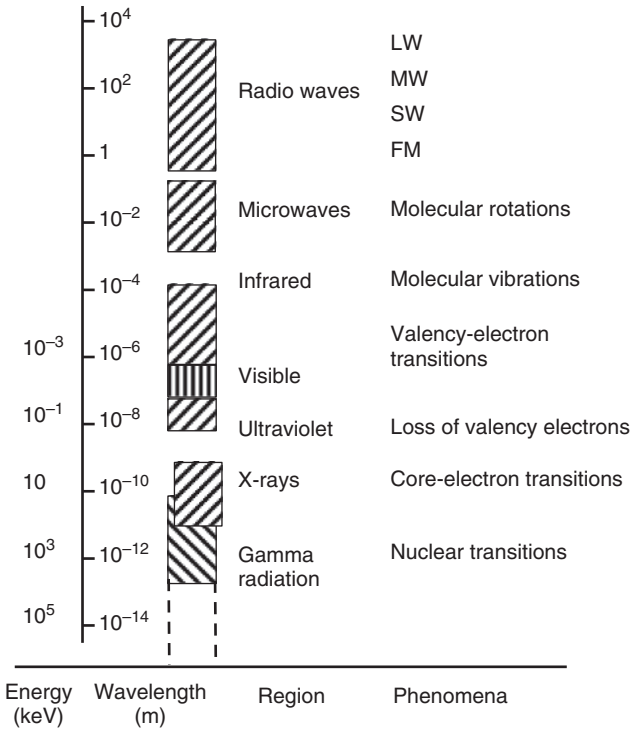


Figure 1.14 The electromagnetic spectrum.

Wavelength, λ, and frequency, ν, are, in principle, equally valid as energy units for characterizing these radiations, and indeed are the preferred units in other parts of the electromagnetic spectrum. Relationships between these quantities for all electromagnetic radiation are as follows:

$$E = h \times \nu \tag{1.1}$$

and

$$\lambda \times \nu = c \tag{1.2}$$

where h (the Planck constant) $= 4.135 \times 10^{-15}$ eV s and c (the velocity of light, or any electromagnetic radiation, in a vacuum) $= 2.997\,926 \times 10^8$ ms^{-1}. Thus, 1000 keV corresponds to a wavelength 1.2398×10^{-12} m, or a frequency of 2.4180×10^{20} Hz. There is some overlap between higher energy X-rays (X-rays range is from just below one to just above 100 keV) and lower energy gammas (whose range we will assume here to be from 10 to 10 000 keV). The different names are used merely to indicate different origins.

The 10^5 keV in the figure is by no means the upper limit to energy. Astronomers detect so-called 'cosmic gamma-rays' (more strictly photons) at much higher energies. Our common energies of around 10^6 eV would be their 'soft' gammas. Above that is 'medium energy' to 3×10^7 eV, 'high energy' to 10^{10} eV, 'very high energy' to 10^{13} eV and 'ultra-high energy' to $>10^{14}$ eV. Measurement of the higher energies is via the interaction of secondary electrons, which are produced in the atmosphere; large-scale arrays of electron detectors are used.

We have already seen that gamma emissions are the result of transitions between the excited states of nuclei. As the whole technique of gamma spectrometry rests on (1) the uniqueness of gamma energies in the characterization of radioactive species and (2) the high precision with which such energies can be measured, it is of interest to consider briefly some relevant properties of the excited states.

1.6.2 Some Properties of Nuclear Transitions

It is sometimes useful to think of nucleons in a nucleus as occupying different energy levels in much the same way as electrons are arranged in shells outside the nucleus. Then, exactly as quantum theory predicts that only particular electron energies are available to extranuclear electrons giving K, L, M shells, etc. so calculations for the nucleus only allow the occupation of certain energy levels for neutrons and, independently, for protons. An excited nuclear state occurs when one or more nucleons have jumped up to a higher energy level or some (or all nucleons) move coherently as a vibration or rotation. Our interest here is in the movement between excited states and in what controls the probability of this occurring.

Nuclear energy states vary as charge and current distributions in the nucleus change. Charge distributions result in electric moments; current distributions give rise to magnetic moments (the neutron may be uncharged, but it still has a magnetic moment). Consider first the electric moment. Oscillating charges can be described in terms of spherical harmonic vibrations, which may be expressed in a multipole expansion. Successive terms in such an expansion correspond to angular momenta in definite quantized units. If one unit of angular momentum is involved, this is called electric dipole radiation and indicated by E1; if two units are involved, we have electric quadrupole radiation, E2 and so on. Likewise, there is a parallel system of magnetic multipoles corresponding to changes in magnetic moments, which give rise to M1 for the magnetic dipole, M2 for the magnetic quadrupole, etc.

As well as changes in angular momentum, there is also the possibility of a change in parity, π. This concept is a property of wave functions and is said to be either + or − (even or odd), depending on the behaviour of the wave function as it is mathematically reflected in the origin. So, there are three properties of a nuclear transition:

- Is it an electric or magnetic transition, E or M?
- Which multipolarities are involved, or what is the change in angular momentum, e.g. E1, E2, E3?
- Is there a change of parity?

These ideas are used in formulating selection rules for gamma transitions. This gives a sound theoretical basis to the apparently arbitrary probability of the appearance of particular gamma emissions. Sometimes, decay schemes have energy levels labelled with spin and parity, as well as energy above the ground state. Figure 1.15 shows examples of this, with the type of multipole transitions expected according to the selection rules.

1.6.3 Lifetimes of Nuclear Energy Levels

Nuclear states also have definite lifetimes, and where transitions would involve a large degree of 'forbiddenness' according to the selection rules, the levels can be appreciably long-lived. If the lifetime is long enough to be easily measurable, then we have an **isomeric state**. The half-life of the transition depends on whether it is E or M, on the multipolarity, on the energy of the transition and on the mass number. Long half-lives are strongly favoured where there is high multipolarity (e.g. E4 or M4) and low transition energy. Most gamma transitions occur in less

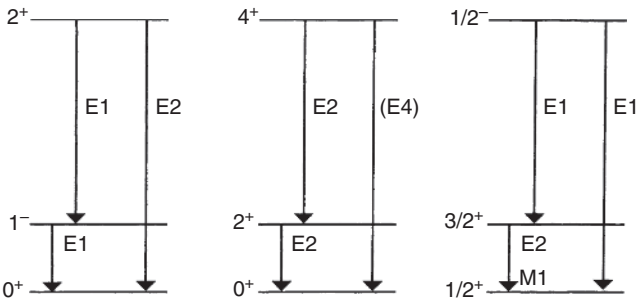


Figure 1.15 Representation of some gamma decay schemes showing spins, parities and expected multipole transitions.

than 10^{-12} s. As to what is readily measurable in practice is something of a moot point, but certainly milliseconds and even microseconds give no real problems. Some would take 1 ns as the cut-off point.

These nuclear isomers, sometimes said to be in **metastable states**, are indicated by a small 'm' as superscript. An example is the 661.7 keV level in ^{137}Ba (see Figure 1.7); this has a half-life of 2.552 m and would be written as ^{137m}Ba (sometimes seen as ^{137}Ba^{m} and deprecated by these authors). Note that in the measurement of ^{137}Cs there is no indication that this hold-up in the emission process exists. Only a rapid chemical separation of barium from caesium, followed by a count of the barium fraction, would show the presence of the isomer. Normally, the 661.7 keV gamma-ray appears with the half-life of the ^{137}Cs because ^{137}Cs and ^{137m}Ba are in secular equilibrium (see Section 1.8.3).

Some half-lives of isomeric states can be very long, for example, ^{210m}Bi decays by alpha emission with a half-life of 3.0×10^{6} a. Alpha decay is, however, a rare mode of decay from a metastable state; gamma-ray emission is much more likely. A gamma transition from an isomeric state is called an **isomeric transition** (IT). On the Karlsruhe Nuclide Chart, these are shown as white sections within a square that is coloured (if the ground state is radioactive) or black (if the ground state is stable).

1.6.4 Width of Nuclear Energy Levels

A nuclear energy level is not at an infinitely precise energy but has a certain finite width. This is inversely related to the lifetime of the energy level through the Heisenberg Uncertainty Principle, which may be expressed as follows:

$$\delta E \times \delta t \geq h/2\pi \, (= 6.582\,122 \times 10^{-16} \text{ eV s}) \tag{1.3}$$

where,

- δE is the uncertainty in the energy, which we will assume to be equivalent to an energy resolution (expressed as full width of the peak at half maximum height (FWHM)).
- δt is the uncertainty in time, taken as the mean lifetime of the level; mean lifetime being $1/\lambda$, where λ is the decay constant, or alternatively $1.4427 \times t_{1/2}$, where $t_{1/2}$ is the half-life.
- h is the Planck constant.

Thus, δE for the 661.7 keV level of ^{137m}Ba whose half-life is 2.552 m will be about 3×10^{-18} eV – exceedingly small. The level involved in the decay of ^{60}Co at 1332.5 keV (see Figure 1.8) has a lifetime of 7×10^{-13} s; this implies an energy width of about 9×10^{-4} eV. This is still very small compared to the precision with which gamma energies can be measured and with the FWHM of spectrum peaks, typically 1.9 keV at 1332.5 keV. In general, the widths of the nuclear energy levels involved in gamma emission are not a significant factor in the practical determination of gamma energies from radioactive-decay processes. This is considered further in Chapter 6.

1.6.5 Internal Conversion

The emission of gamma radiation is not the only possible process for de-excitation of a nuclear level. There are two other processes: internal conversion (IC) and pair production.

Pair production as a form of de-excitation is uncommon, and I will only touch upon it here. There are close similarities with the process, which will be described in detail in Section 2.2.3 on interactions of gamma radiation with matter, where pair production is, by contrast, of major importance. It is only possible if the energy difference between levels is greater than 1022 keV, when that part of the total energy is used to create an electron–positron pair. These two particles are ejected from the nucleus and share the remainder of the decay energy as kinetic energy. An example of decay by pair production is the isomeric transition of ^{16m}O, which has a half-life of 7×10^{-11} s and a decay energy of 6050 keV.

Internal conversion, on the other hand, is very common. In this process, the energy available is transferred to an extranuclear electron, which is ejected from the atom. This is called an **internal conversion electron**.

It is mono-energetic, having an energy equal to the transition energy less the electron binding energy and a small nuclear recoil energy. Measurement of the distribution of electron energy (i.e. an electron spectrum) would reveal peaks corresponding to particular electron shells, such as K, L and M. Loss of an electron from a shell leaves a vacancy, and this vacancy will be filled by an electron dropping into it from a higher shell. Thus, as with electron capture, an array of X-rays and Auger electrons will also be emitted.

However, note that because IC is a mode of de-excitation and there is no change in Z, N or A, the X-radiation that is produced is characteristic of the *parent* nuclide. Both 'parent level' and 'daughter level' are the same element. This is in contrast with electron capture, where the X-rays are characteristic of the daughter. If X-ray energies are to be used as a diagnostic tool, the user must know which decay process is occurring.

Internal conversion operates in competition with gamma-ray emission, and the ratio of the two is the **internal conversion coefficient**, α:

$$\alpha = \frac{\text{number of IC electrons emitted}}{\text{number of gamma-rays emitted}} \tag{1.4}$$

This may be subdivided into α_K, α_L, etc. where electrons from the individual K and L shells are considered. Values of α depend on the multipolarity, transition energy and atomic number. In broad terms, α increases when half-life and Z increase and as ΔE decreases. At high Z, transitions with small transition energies may be 100% converted.

A practical point arises regarding the use of information taken from decay scheme diagrams. It cannot be assumed that because x% of disintegrations are feeding a certain energy level, then the same x% of disintegrations will produce a gamma-ray of that energy. An example of this is the decay of ^{137}Cs. The decay scheme (see Figure 1.7) shows that 93.5% of decays populate the 661.7 keV level in the daughter. However, tables of decay data state that the emission probability of the 661.7 keV gamma is only 85.1%. Thus, 8.4% of the gamma decays are internally converted; a number which could be calculated from the coefficient α. Another example is the decay of ^{228}Th. It may be deduced from Figure 1.13 that some 27% of the α decays feed the ^{224}Ra level at 84.4 keV, yet the emission probability of the 84.4 keV gamma-ray is only 1.2%, and instead many Ra X-rays are emitted as a consequence of Internal conversion.

1.6.6 Abundance, Yield and Emission Probability

It is common for the number of gamma-rays emitted by a nuclide to be referred to as 'the abundance' and sometimes as the 'yield'. Both of these terms lack precision. Historically, confusion was often caused because an author or data source would quote abundances that were effectively beta transition data – the 93.5% figure quoted earlier. In fact, the proportion of decays that give rise to 661.7 keV gamma-rays in the aforementioned example is 85.1% when internal conversion is taken into account. In this text, I will use the term **gamma emission probability** on the basis that it says exactly what it means – the probability that a gamma-ray will be emitted, all other factors being taken into account.

1.6.7 Ambiguity in Assignment of Nuclide Identity

We have seen how gamma-rays are emitted with very precisely defined energies; these energies being characteristic of particular radionuclides. The majority of ambiguities that arise in allotting nuclides to energies would probably be overcome if the energy resolution of detector systems were improved. However, there is a not uncommon situation where discrimination between radionuclides by gamma energy alone is in principle not possible. This is where isobaric nuclides are decaying to the same stable product from either side. The gamma radiation is the result of transitions within energy levels in the stable nuclide; there can only be one set of energy levels and thus the gamma energies must be the same, regardless of how the energy levels are fed.

Figure 1.16 shows the region of the (Karlsruhe) Chart of the Nuclides showing how both ^{51}Cr and ^{51}Ti decay to ^{51}V. The relevant decay schemes are summarized in Figure 1.17. Data compilations give gamma energies and emission probabilities, as shown in Table 1.2. There is no way of distinguishing between the major gamma energies

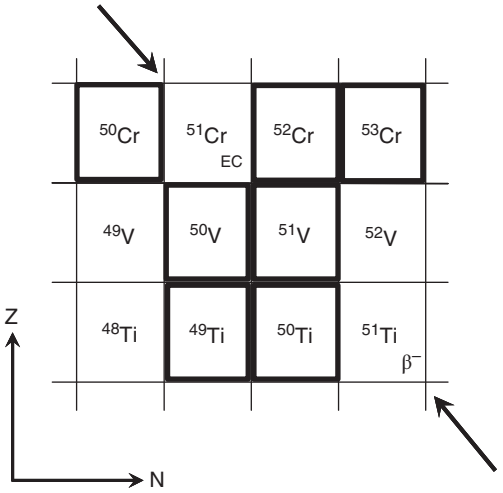


Figure 1.16 Chart of the Nuclides, showing part of the isobar $A = 51$. Heavy boxes indicate the stable nuclides.

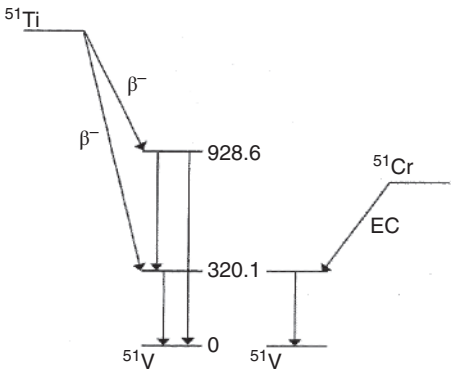


Figure 1.17 Decay schemes of ^{51}Ti and ^{51}Cr.

Table 1.2 Gamma energies shown by two isobaric nuclides.

Nuclide	Gamma energy (keV)	Emission probability, P_γ
^{51}Ti	320.084	0.931
	608.55	0.0118
	928.63	0.069
^{51}Cr	320.084	0.0987

at 320 keV as they are identical. The situation in this case is of practical interest as both ^{51}Ti and ^{51}Cr are the only gamma-emitting thermal neutron activation products of these particular elements. If there were sufficient activity present, then the other lower emission probability gammas from ^{51}Ti should be visible; so allowing discrimination from ^{51}Cr, but if there is only a small peak at 320 keV, these other gamma peaks may not be apparent. ^{51}Cr, as an EC nuclide, emits X-rays; these are of vanadium, but with energies at 4.95 and 5.43 keV may well be below the energy range of the detector. The other nuclear parameter is half-life, and in this case, the half-lives are very different, i.e. ^{51}Ti, 5.76 m and ^{51}Cr, 27.71 d. However, a single count does not give any half-life information.

1.7 Other Sources of Photons

1.7.1 Annihilation Radiation

In Section 1.2.2, I explained how positron decay gives rise to an annihilation peak at 511.00 keV, and how this is distinguishable by being much broader than would be expected. The Doppler effect could add 2 keV to other uncertainties contributing to the width of spectrum peaks. There are reports of very small variations in energy (about 1 in 10^5) depending on the atomic mass of the material; this will be of no practical significance. Some positron emitters, such as ^{22}Na, have been used as energy calibration standards and ^{22}Na, in particular, has the advantage of a very simple spectrum with only two widely spaced peaks. However, care must be taken if the calibration program also takes the opportunity of measuring peak width or peak shape, for which the 511 keV peak is quite unsuited. In general, the annihilation peak must be regarded as a special case needing some thought in its interpretation. Further information is given in Sections 2.2.3 and 2.5.3.

1.7.2 Bremsstrahlung

Bremsstrahlung is a German word meaning 'slowing-down radiation'. It is electromagnetic radiation produced by the interaction of fast electrons with the Coulombic field of the nucleus. The electron energy loss appears as a continuum of photons, largely noticed in the X-ray region, although in principle, the maximum energy is that of the beta particle. Other energetic particles lose energy in a similar way, but bremsstrahlung is only significant with light particles, the effect being inversely proportional to the square of the charged particle mass. The effect on a gamma spectrum is to raise the general background continuum, hence making the detection of a superimposed gamma-ray more difficult. There is an inverse relationship between the number of quanta emitted and the photon energy, so that the bremsstrahlung background level decreases with increasing energy.

There is a larger bremsstrahlung interaction with higher atomic number absorbers and higher electron (beta) energies. In practice, for 1000 keV beta particles in lead ($Z = 82$), there is an appreciable effect; for 1000 keV beta particles in aluminium ($Z = 13$), the effect is unimportant. It follows that any structure near the detector, a sample jig, for example, should be constructed of a low-Z material such as a rigid plastic. The use of a graded shield (see Section 2.5.1) will minimize bremsstrahlung in the same way. In complete contrast, the effect is put to positive use when a source of high intensity 'X-radiation' is required for photon activation analysis or medical purposes.

There are occasional reports of attempts to reduce the bremsstrahlung effect in gamma spectra by using an electromagnetic field near the source to divert betas away from the detector. This is clearly a cumbersome procedure and has been found to be of limited value in practice.

1.7.3 Prompt Gamma-Rays

These are gamma-rays emitted during a nuclear reaction. If we consider the thermal neutron activation of cobalt, which in the short notation is expressed as follows:

$$^{59}\mathrm{Co}(\mathrm{n},\gamma)^{60}\mathrm{Co}$$

Then the gamma-ray shown is a prompt gamma released as the excited ^{60}Co nucleus falls to the ground state. This happens quickly, in less than 10^{-14} s, is a product of a nuclear excitation level of ^{60}Co itself and unrelated to the gamma emissions of the subsequent radioactive decay of the ^{60}Co, which as we have seen is a property of ^{60}Ni. Any measurements of prompt gammas must necessarily be made online, and special equipment is needed, for example, to extract a neutron beam from a reactor. Energies are often greater than those of beta decay gammas, going up to more than 10 MeV. Analytically, the method is useful for some low-Z elements, which do not give

activation products with good decay gammas. Elements, such as H, B, C, N, Si, P and Ca, have been determined by such means. I will discuss later how prompt gammas can appear from detector components if systems are operated in neutron fields (Chapter 11), even very low naturally occurring neutron fields.

1.7.4 X-rays

I have described how X-radiation appears as a result of the rearrangement of the extranuclear atomic electrons after electron capture and internal conversion. X-rays are mono-energetic, the energy being equal to the difference between electron energy levels (or very close to this: conservation of energy and momentum mean that a very small recoil energy must be given to the whole atom, thus reducing the X-ray energy slightly).

1.7.4.1 X-ray Nomenclature

X-radiation has been known from the very earliest days of nuclear science; Röntgen named the rays in 1895, before Becquerel's discovery of radioactivity, and the classification of X-rays inevitably proceeded in a piecemeal fashion. Hence, while Figure 1.18(a) is the sort of logical nomenclature one might pursue with today's hindsight, in practice, this is only an approximation to the complicated real situation of Figure 1.18(c). Broadly, K_α, K_β, L_α and L_β fit the format more or less correctly, but the logical system fails even with some of these and does so most decidedly with more distant transitions.

The K shell has only one energy level. Higher shells have sub-shell levels; they are said to be degenerate. This means that the electrons in each shell, apart from the K shell, do not have exactly the same energy. There is *fine structure* present, giving s, p, d, etc. sub-shells, and as exemplified in Figure 1.18(b), this gives rise to fine structure in the X-rays; the sub-divisions are labelled as $K_{\alpha1}$, $K_{\alpha2}$, $L_{\gamma6}$ and so on. Not all possible transitions occur in reality because selection rules operate based on angular momentum changes. Thus, the transition from L_1 to K is very unlikely to occur and said to be 'hindered'.

1.7.4.2 X-ray Energies

Present-day germanium detectors have sufficient resolution to separate the energies involved in the fine structure of elements with high values of Z, where the differences are marked, but would not resolve fine structure at low Z. Because of the widespread use of lead as a shielding material, that particular set of X-rays occurs very commonly

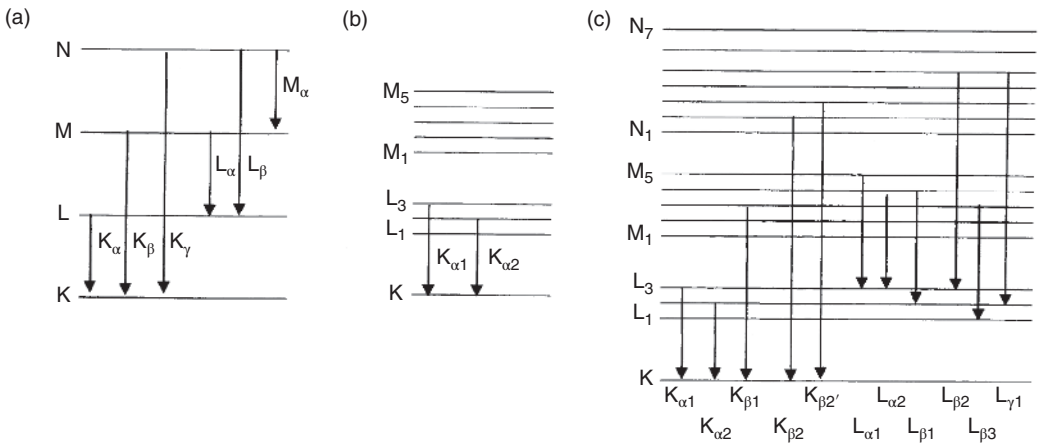


Figure 1.18 X-ray nomenclature: (a) how in an ideal world, the broad groups of X-rays should be named (see text); (b) an example of fine structure; (c) how in the real world, some X-rays are named.

Table 1.3 Some K X-rays.

Element	Z	X-ray energy (keV)			
		$K_{\alpha 1}$	$K_{\alpha 2}$	$K_{\beta 1}$	$K_{\beta 2}$
Copper	29	8.047	8.027	8.904	8.976
Germanium	32	9.885	9.854	10.981	11.10
Cadmium	48	23.174	22.984	26.084	26.644
Lead	82	74.969	72.805	84.784	97.306
Uranium	92	98.434	94.654	111.018	114.456

in gamma spectra. L-series and M-series X-radiation is much lower in energy than the K-series shown. A comprehensive listing of energies is given by Browne and Firestone (1986); a few representative values are shown in Table 1.3.

1.7.4.3 X-rays and Identification

It should be noted that X-ray measurements can give information as to the element involved but will not identify the isotope of the element. This is because the arrangement of the atomic electrons is determined only by the number of protons in the nucleus (Z) and not by its mass (A). I repeat the point made earlier, that in order to identify a radioactive element from X-ray energies, we need to know the type of decay involved. Only in internally converted isomeric transitions are the X-ray characteristic of the radionuclide itself. In electron capture they identify the daughter; if the daughter has atomic number Z, then that of the decaying nuclide is $Z + 1$.

1.7.4.4 The Energy Widths of X-rays

We have seen how nuclear energy levels have very small widths, which have negligible impact on the width or shape of gamma peaks in spectra. This is not always the case with X-rays. The width of atomic electron levels is described by a Lorentzian function (as are nuclear levels) and this contains a width parameter Γ similar to an FWHM (see Section 6.1). The Lorentzian function is

$$L(x) = \frac{\Gamma/2\pi}{(x - x_0)^2 + (\Gamma/2)^2} \tag{1.5}$$

while the Gaussian function is

$$G(x) = \frac{1}{\sigma \times \sqrt{2\pi}} \exp \frac{-(x - x_0)^2}{2\sigma^2} \tag{1.6}$$

and FWHM $= 2.355 \times \sigma$.

Figure 1.19 shows both functions, drawn so as to make Γ = FWHM. Peak broadening in the detection process is described by Gaussian functions (Chapter 6). The pulse size distribution going through the detection system reflects both the initial Lorentzian energy distribution and the imposed Gaussian broadening. A convolution of these two, in practice usually with wildly differing 'widths', gives a **Voight function**.

An X-ray energy is the difference between an initial and a final state, and so the intrinsic width of both levels must be taken into account. With Lorentzian functions, these are simply added directly. Table 1.4 shows the intrinsic width of some atomic electron levels and the corresponding X-ray and compares these to the actual X-ray energy and the resolution of a good detector at that energy. It can be seen that

- the intrinsic width of electron energy levels (column (b)) is many orders of magnitude greater than the width of nuclear levels (typical examples in Section 1.6.4 are 3×10^{-18} and 9×10^{-4} eV);

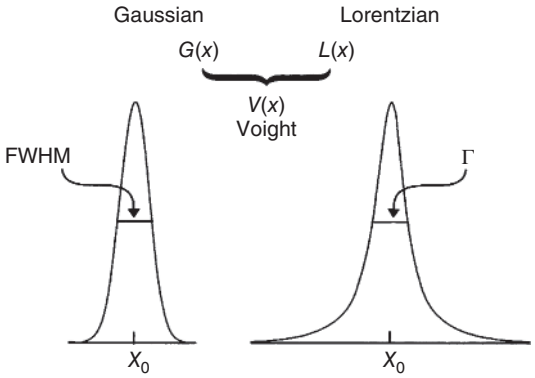


Figure 1.19 The Lorentzian and Gaussian distributions, drawn with the same x-axes and with Γ = FWHM. A convolution of these gives a Voight function.

Table 1.4 The effect of the intrinsic width of atomic energy levels[a),b)].

Element	Z	(a) $K_{\alpha 2}$ X-ray energy (keV)	(b) Width of level (eV)		(c) Width of $K_{\alpha 2}$ X-ray (eV)	(d) $\frac{\text{Width } K_{\alpha 2}}{\text{Energy } K_{\alpha 2}}$	(e) FWHM of good detector (eV)	(f) $\frac{\text{Width } K_{\alpha 2}}{\text{FWHM}}$
			K	L_2				
Nickel	28	7.45	1.44	0.52	1.96	0.000 26	155	0.013
Cadmium	48	22.98	7.28	2.62	9.9	0.000 42	200	0.049
Lead	82	72.80	60.4	6.5	66.8	0.000 92	350	0.19
Uranium	92	94.65	96.1	9.3	105.4	0.0011	415	0.25

a) Level width data from Krause and Oliver (1979), *J. Phys. Chem. Ref. Data*, **8**, 329.
b) Detector resolution assumed to vary linearly from 150 eV at 5.9 keV to 500 eV at 122 keV.

- the width of the X-ray is the sum of the widths of the two relevant energy levels (Columns (b) and (c));
- the widths are a strong function of Z;
- intrinsic X-ray widths are small (<0.1%) compared with X-ray energies, even at high Z values (Column (d));
- intrinsic X-ray widths have a considerable influence on the widths of their peaks in spectra at higher Z values (Column (f)). The ratios in this column can be read as some measure of the proportions of Lorentzian and Gaussian in the mixed Voight function. More pertinently, the Lorentzian function has long tails (Figure 1.19) and a significant contribution from this intrinsic line shape will lead to peak shapes that also have long tails. This is known as **Lorentzian broadening** and may well affect computer analysis of peak areas. If counts in the tails are not included, then areas of high-Z X-rays will be underestimated by up to a few percent.

1.8 The Mathematics of Decay and Growth of Radioactivity

Radionuclides are, by definition, unstable and decay by one, or more, of the decay modes: alpha, beta-minus, beta-plus, electron capture or spontaneous fission. Although strictly speaking a de-excitation rather than a nuclear decay process, we can include isomeric transition in that list from the mathematical point of view. The amount of a radionuclide in a sample is expressed in **Becquerels** – numerically equal to the rate of disintegration – the number of disintegrations per second. We refer to this quantity as the **activity** of the sample. The activity will change with time so we must always specify at what time the activity was measured.

From time to time, we will have to take into account the fact that one radionuclide will decay into another, but for the moment, we can consider only the simple decay process.

1.8.1 The Decay Equation

Radioactive decay is a first-order process. The rate of decay is directly proportional to the number of atoms of radionuclide present in the source, i.e. the activity, A, is directly proportional to the number of atoms, N, of nuclide present:

$$A = \mathrm{d}N/\mathrm{d}t = -\lambda N \tag{1.7}$$

The proportionality constant, λ, is called the **decay constant** and has the units of reciprocal time (e.g. s^{-1}, h^{-1}). The reciprocal of the decay constant is the **mean lifetime**, τ, of the radionuclide, the average time with which an atom can be expected to exist before its nucleus decays:

$$\tau = 1/\lambda$$

This time represents a decay of the source by a factor of e (i.e. 2.718). It is more convenient and meaningful to refer to the **half-life**, $t_{1/2}$, of the radionuclide – the time during which the activity decreases to half its original value:

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

For example, for ^{60}Co,

Decay constant: $3.60 \times 10^{-4}\ \mathrm{d}^{-1}$
Half-life: 1925.5 ± 0.5 d (5.27 a)
Mean lifetime: 2777.9 d

Equation (1.7) leads to the more commonly used decay equation relating number of atoms (N_t) at time (t) and half-life ($t_{1/2}$):

$$N_t = N_0 \exp(-\ln 2 \times t/t_{1/2}) \ \text{ or } \ N_t = N_0 \exp(-\lambda t) \tag{1.8}$$

where N_0 is the number of atoms at time $t = 0$. In practice, it is more useful to replace number of atoms by activity, bearing in mind that activity is proportional to the number of atoms:

$$A_t = A_0 \exp(-\lambda t) \tag{1.9}$$

Figure 1.20(a) illustrates the shape of the decay curve. If we take logarithms of our activity (Equation (1.9)), we transform it into a linear relationship:

$$\log A_t = \log A_0 - \lambda t$$

So, when plotted on a logarithmic scale, the activity of a source over a period of time would be a straight line (Figure 1.20(b)). Indeed, if a straight line is not obtained, then one can be sure that more than one nuclide is being measured or an isomeric state may be present. In favourable cases, it is possible to resolve composite decay curves to estimate the relative amounts of the component radionuclides and their half-lives. (Resolution is easier, of course, if the half-lives are known.)

1.8.2 Growth of Activity in Reactors

Most radionuclides are created by nuclear reactions. A typical example would be neutron activation within a nuclear reactor. The activity at a point in time after the start of irradiation represents a balance between the rate of creation of radioactive atoms and the rate of decay, which increases as the number of atoms increases. The rate of growth of activity can be expressed as follows:

$$\mathrm{d}N/\mathrm{d}t = N_\mathrm{T}\sigma\phi$$

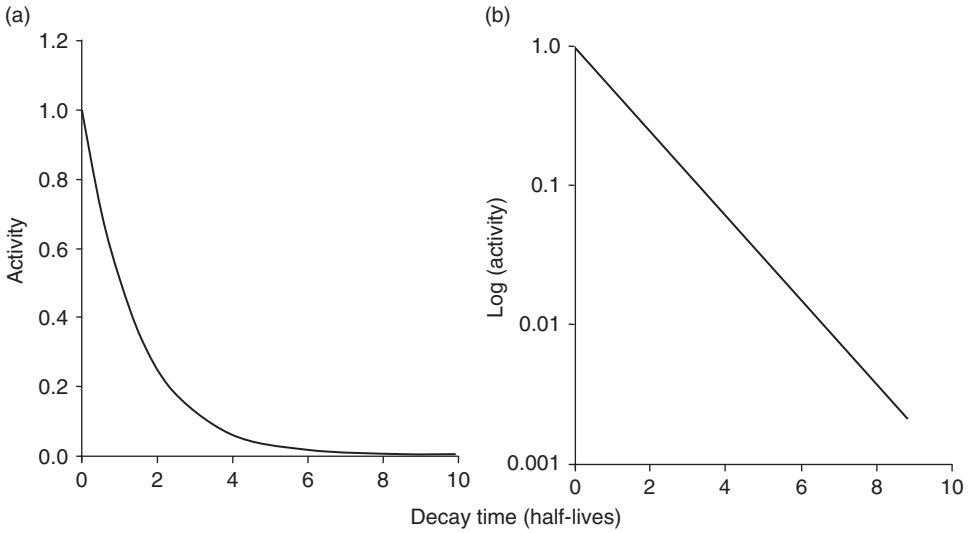


Figure 1.20 The decay of a radioactive nuclide: (a) linear scale; (b) logarithmic scale (activity is plotted relative to that at the start of the decay).

where N_T is the number of target atoms, ϕ is the neutron (or, in general, the particle) flux and σ is the cross section for the reaction, which we might imagine in this case to be that for thermal neutron capture, (n, γ), reaction. Within a particular system all these terms are constant (or at least they are until a significant fraction of target atoms are 'burned up'). This means that, in the short term, the rate of growth of activity is constant.

The rate of decay is governed by Equation (1.7) and increases as the number of atoms of radionuclide increases. Common sense suggests that the rate of decay can never be greater than the rate of growth and that at some point in time the rate of decay must become equal to the rate of growth. Solving the resulting differential equation leads to the following:

$$A_t = A_S[1 - \exp(-\lambda t)] \tag{1.10}$$

where t is the time of irradiation and A_S is the **saturation activity** – the maximum activity that can be achieved during an irradiation. This is illustrated in Figure 1.21. The activity rises to within 0.1% of saturation after 10 half-lives of the product. A simple practical consequence of the growth equation is that short-lived nuclides reach saturation very quickly, long-lived nuclides very slowly.

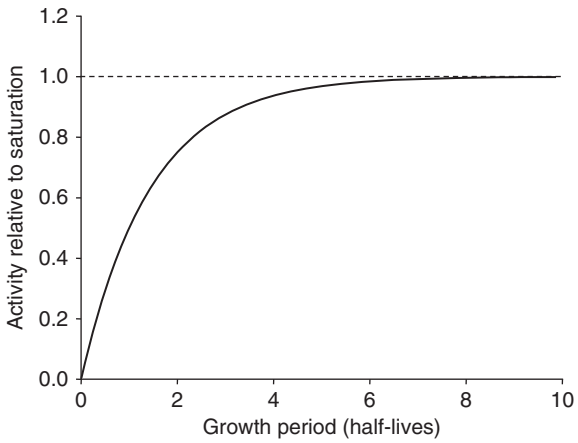


Figure 1.21 The growth of radioactivity in a nuclear reactor.

1.8.3 Growth of Activity from Decay of a Parent

When one radionuclide (the parent) decays into another radionuclide (the daughter), the rate of change of the number of daughter atoms must be the difference between the rate of growth from the parent and the rate of decay of the daughter:

$$\frac{dN_D}{dt} = \lambda_P N_P - \lambda_D N_D$$

$$\frac{dN_D}{dt} = \lambda_P N_{P0} \exp(-\lambda_P t) - \lambda_D N_D$$

where the subscripts D and P refer to daughter and parent, respectively, and the subscript 0 indicates the number of atoms at $t = 0$. Solving this linear differential equation gives the following:

$$N_D = \frac{N_{P0}[\exp(-\lambda_P t) - \exp(-\lambda_D t)]\lambda_P}{(\lambda_D - \lambda_P)} + N_{D0} \exp(-\lambda_D t) \tag{1.11}$$

Bearing in mind that $A = \lambda N$, we can rewrite this in terms of activity:

$$A_D = \frac{A_{P0}[\exp(-\lambda_P t) - \exp(-\lambda_D t)]\lambda_P}{(\lambda_D - \lambda_P)} + A_{D0} \exp(-\lambda_D t) \tag{1.12}$$

To gain an understanding of this relationship as the ratio of parent to daughter half-lives varies, it is useful to imagine that we chemically separate the parent and daughter, in which case the second term of Equation (1.12) is zero. We can then follow the change in their activities in the initially pure parent and calculate the total activity. There are three particular cases depending upon the relation between the parent and daughter half-lives.

1.8.3.1 Transient Equilibrium – $t_{1/2}$ Parent > $t_{1/2}$ Daughter

In a **transient equilibrium**, the activity of the daughter nuclide is in constant ratio to that of the parent nuclide and apparently decays with the half-life of the parent. In Figure 1.22, we see the decay of the parent unaffected by the absence or presence of daughter and the growth of the daughter activity. (The timescale is in units of half-life of the daughter nuclide.) The daughter grows faster than decay of the parent and being of shorter half-life eventually exceeds the activity of the parent. The total activity in the system is the sum of the parent and daughter activities. Transient equilibrium is established after about 10 half-lives of the daughter nuclide after which the daughter apparently decays with the half-life of the parent.

This should not be forgotten when measuring nuclides that are sustained by decay of a parent. For example, if ^{95}Nb is measured in a fission product mixture, it is likely, using the commonly available commercial gamma spectrum analysis packages, that the normal half-life of ^{95}Nb, 34.98 d, will be extracted from the package's nuclide library and used to correct ^{95}Nb activities. In fact, if the age of the fission product mixture is greater than a year or so, then the appropriate half-life is that of the parent ^{95}Zr, i.e. 64 d. At shorter decay times, the accurate calculation of ^{95}Nb activity will be beyond the capabilities of a standard analysis program. Fission product mixtures contain many isobaric decay chains and many such parent/daughter pairs of nuclides; this problem is not at all uncommon.

If we take Equation (1.12) and set t to a value much greater than the half-life of the daughter, we can calculate the relative numbers of parent and daughter atoms at equilibrium:

$$N_D/N_P = \lambda_P/(\lambda_D - \lambda_P) \tag{1.13}$$

And the equilibrium activity of the daughter relative to that of the parent:

$$A_D = A_P \lambda_D/(\lambda_D - \lambda_P) \text{ or, in terms of half-lives}$$

$$A_D = A_P t_{1/2P}/(t_{1/2P} - t_{1/2D}) \tag{1.14}$$

where $t_{1/2P}$ and $t_{1/2D}$ are the half-lives of parent and daughter, respectively.

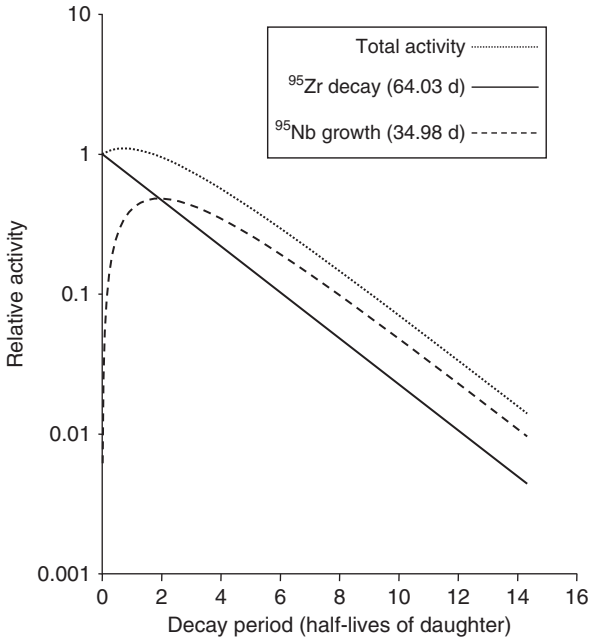


Figure 1.22 Transient equilibrium – relative activities of parent and daughter nuclides after separation.

Figure 1.22 also shows that, as equilibrium is approached, the activity of the daughter nuclide becomes greater than that of the parent. This may, at first sight, seem odd. Take the case of ^{140}La in equilibrium with its parent ^{140}Ba (half-lives of 1.68 and 12.74 d, respectively) and apply the aforesaid equations. At equilibrium, the number of ^{140}La atoms in the source will be only 0.15 times the number of ^{140}Ba atoms. However, because ^{140}La has a shorter half-life, its activity will be 15% greater than that of the parent ^{140}Ba. The difference can be substantial; in an equilibrium mixture of ^{95}Zr/^{95}Nb, the activity of the ^{95}Nb is 64/(64 – –35), i.e. 2.2 times that of the ^{95}Zr.

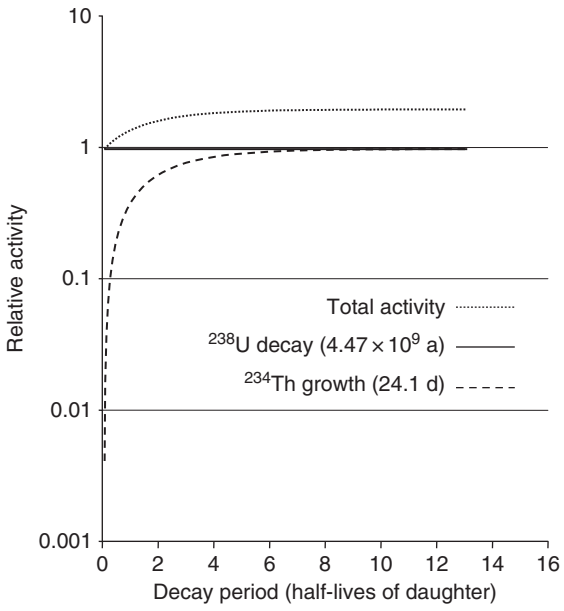


Figure 1.23 Secular equilibrium – relative activities of parent and daughter nuclides after separation where the parent half-life is much greater than that of the daughter.

1.8.3.2 Secular Equilibrium – $t_{1/2}$ Parent $\gg$ $t_{1/2}$ Daughter

If the half-life of the parent nuclide is very long compared with that of the daughter, the equilibrium state is referred to as **secular equilibrium**. In such situations, where $t_{1/2D}$ becomes negligible, Equation (1.14) becomes $A_D = A_P$, i.e. the daughter activity equals the parent activity (Figure 1.23).

Take, for example, the first three stages of the ^{238}U decay series shown in Figure 1.24. We find that in each parent/daughter case, the half-life of the parent is very much greater than the daughter and therefore there will be a secular equilibrium established between each pair. The activity of each daughter will be equal to that of its parent and the total activity, for this portion of the decay chain, will be three times that of the ^{238}U. Note that as far as the ^{234}Pa and the ^{234m}Pa are concerned, at equilibrium, the half-life of their parent is effectively that of ^{238}U. The branching of the ^{234}Th decay means that the total ^{234}Pa + ^{234m}Pa will be, at equilibrium, equal to that of the ^{238}U. The activity will be shared between the two nuclides according to the branching ratio.

If we look at the complete ^{238}U decay scheme, we find 14 daughter nuclides, all of whom have much shorter half-lives than their ultimate parent. The total activity, assuming radioactive equilibrium is established, will be 14 times the ^{238}U activity. (A more complete discussion of gamma spectrometry of the uranium and thorium decay series nuclides can be found in Section 14.2.)

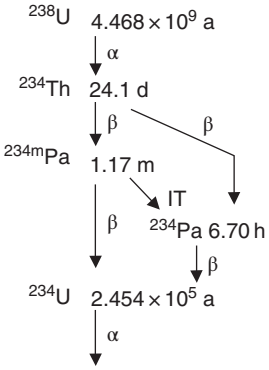


Figure 1.24 The first few stages of the ^{238}U decay series (IT is isomeric transition).

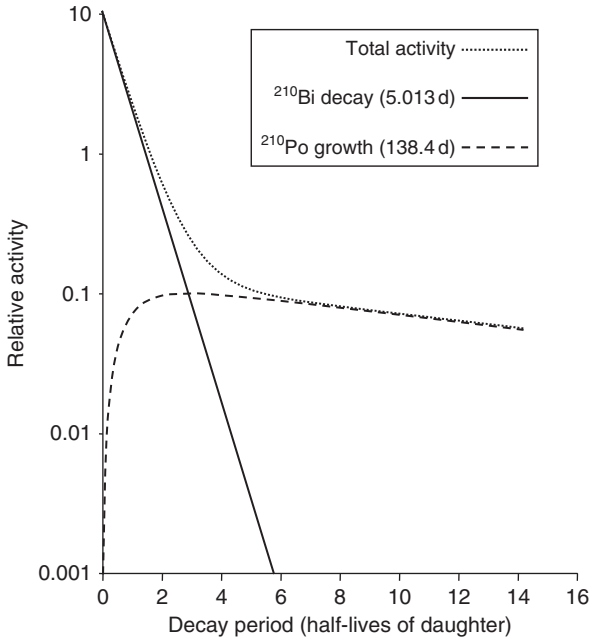
1.8.3.3 No Equilibrium – $t_{1/2}$ Parent < $t_{1/2}$ Daughter

If the daughter half-life is greater than that of the parent then, obviously, the parent will decay, leaving behind the daughter alone. Figure 1.25 shows the growth of daughter activity within an initially pure parent. No equilibrium is established; ultimately the decay curve will be that of the grown-in daughter.

1.8.3.4 Multiple Parent–Daughter Relationships

In Chapter 14, we discuss the decay chains of the naturally occurring uranium and thorium nuclides. Those involve many nuclides decaying one to another. The mathematics of those relationships follows the principles referred to

Figure 1.25 Relative activities of parent and daughter nuclides after separation where there is no equilibrium.



in Section 1.8.3 but is considerably more complicated. When necessary, the work by Bateman (1910) is still resorted to and there is a reference to it in Further Reading suggestions for this chapter.

1.9 The Chart of the Nuclides

The general layout of the *Karlsruhe Chart of the Nuclides* has been described; it has been used to explain beta decay and parts have been illustrated in Figures 1.3 and 1.16. In this section, I discuss its use in diagnosis, firstly as a data source and then as an indicator of the location of probable nuclides. There are other versions of this chart, but none have made it onto the walls of counting rooms as often as the Karlsruhe Chart.

1.9.1 A Source of Nuclear Data

I would certainly not recommend printed versions of the chart as the best source of nuclear data; the numbers presented are too lacking in detail and (probably) not up to date. There is an interactive online version (see Further Reading) created by the US National Nuclear Data Left – a very useful resource containing much more up-to-date information than the printed versions, but without nuclear cross-section data. The website does, however, have easy links to energy level data and gamma-ray emission data.

The printed chart is nevertheless very useful for rapid assessment. For each element, there is the element symbol, atomic weight and thermal neutron absorption cross section. Within the horizontal strip of isotopes, stable species are shown with a black background and contain the mass number, natural isotopic abundance of the isotope in atom% and thermal neutron absorption cross section, σ, in barns (b). Where appropriate, a stable square will have a white section to indicate the presence of a metastable state of the stable nuclide with its half-life and the energy of the IT gamma decay in keV. Radioactive species are colour-coded: blue, β^-; red, EC or β^+; yellow, α; green, spontaneous fission. If two modes of decay occur, two colours are shown.

For example (referring to Figure 1.26), the square for ^{60}Co would be blue for β^- and contains the half-life, the major maximum beta energies in MeV (useful for bremsstrahlung estimation), major gamma energy (in keV) in order of emission probability and the thermal neutron cross section. The isomer is shown as a white section with decay mode(s) and energies. On this particular chart, electron capture is shown as 'ε', isomeric transition as 'I' and conversion electron emission as 'e'$^-$', along with standard symbols. In Figure 1.26(b), 'σ 20 + 17' indicates 20 b for the (n, γ) cross section to form ^{60m}Co ($t_{1/2}$ = 10.47 m) and 17 b for the (n, γ) reaction to give ^{60}Co directly in its ground state. For most purposes, these numbers would need to be summed if ^{60}Co activity was sought, as essentially all of the metastable states will end up as the ground state within a couple of hours.

The chart contains sufficient information for simple calculations as to whether the quantities of radionuclides found are those that might be expected. The overall activation equation is as follows:

$$A = N_T \times \sigma \times \phi \times [1 - \exp(-\lambda \times t_{irr})] \times \exp(-\lambda \times t_{dk}) \tag{1.15}$$

where

- A = induced activity (in Bq);
- N_T = number of target atoms;
- σ = cross section (probability of reaction taking place; an area, in units of barns (b) where 1 b = 10^{-28} m^2);
- ϕ = flux of activating particle, in most cases, neutrons (units: particles per unit area per second, e.g. n cm^{-2} s^{-1}; note that units of area used must be the same as those used for the cross section);

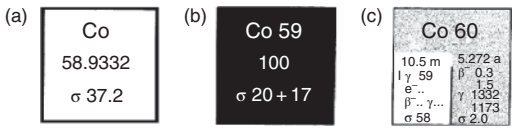


Figure 1.26 Typical data from a chart of the nuclides: (a) the element; (b) a stable isotope; (c) a radioisotope with a metastable state.

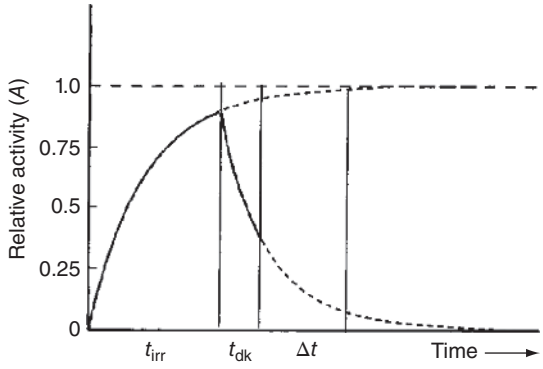


Figure 1.27 The quantity of radionuclide increases during irradiation and then decreases by radioactive decay to the time of measurement: t_{irr}, irradiation period; t_{dk}, decay time; Δt, measurement period.

- t_{irr} = irradiation time (in same units as half-life used to for λ);
- t_{dk} = cooling time (decay time) from the end of the irradiation to the time of measurement (again same units as half-life).

The shape of this expression is shown in Figure 1.27.
The three parts of the equation correspond as follows:

- $[N_0 \times \sigma \times \phi]$ = the saturation activity, indicated by the dashed line at $A = 1.0$; the maximum activity obtainable, asymptotically approached when $t_{irr} > t_{1/2}$.
- $[1 - \exp(-\lambda \times t_{irr})]$ = the approach to saturation; a useful property is that when $t_{irr} = t_{1/2}$, this factor = 0.5, i.e. the activity is half the saturation value; at $t_{irr} = 2 \times t_{1/2}$, the factor is 0.75.
- $[\exp(-\lambda \times t_{dk})]$ = normal radioactive decay from the end of the irradiation to the time of measurement.

1.9.2 A Source of Generic Information

The chart is helpful when tracking down unknown radionuclides detected in a measurement to know what type of nuclear reaction may have been responsible for the production of the activity. In most cases, this will be a thermal neutron reaction resulting in activation by (n, γ), or if the target material is fissile, a fission reaction (n, f).

1.9.2.1 Thermal Neutron Capture (n, γ)

If the activity is due to thermal neutron capture, then the search for nuclides is narrowed dramatically. From more than 2000 radionuclides displayed on the chart, we need to scan only 180 or so. It is clear that as the (n, γ) reaction merely adds one neutron to a stable nuclide we must look at isotopes just one square to the right of a (black) stable one. For example, instead of considering all 21 radioisotopes of arsenic, we need to look at only one; the stable arsenic is ^{75}As and therefore we look at ^{76}As. There are just two qualifications to this simple picture:

- There is one element where there is a significant chance of finding that two neutrons have been added – gold.

^{197}Au + n→	^{198}Au + n→	^{199}Au
stable	β^-	β^-
(σ = 98.8 b)	(σ = 25 100 b)	

This is due to the very large neutron absorption cross section of the first product, ^{198}Au. In most cases, while there may be a considerable amount of activity formed by an (n, γ) reaction, this will correspond to relatively few atoms. Thus, the amount of target material available for a second reaction is small, and a large reaction probability (as we have here with ^{198}Au) is needed to give significant amounts of the second reaction product.

- The second complication is when the (n, γ) product does not decay to a stable nuclide but to a radioactive one. Then nuclides that are an extra transformation away (β^- normally) need to be considered. An example is

^{130}Te(n, γ) →	^{131}Te →	^{131}I →	^{131}Xe
stable	β^- (24 m)	β^- (8.0 d)	stable

or the highly significant sequence,

^{238}U(n, γ) →	^{239}U →	^{239}Np →	^{239}Pu →
'stable'	β^- (23 m)	β^- (2.3 d)	α (2.4×10^4 a)

These transformations can be readily traced on the chart.

A further point: Most elements have more than one stable isotope. So, if one activation product is found, check with the chart for others that could be formed by the same mechanism. For example, if 35.3 h ^{82}Br is seen, formed by (n, γ) from stable ^{81}Br, then look at the chart, which will tell you that there is plenty of stable ^{79}Br with an adequate cross section, so that timescales permitting, there should be 4.4 h ^{80m}Br present and its daughter ^{80}Br as well.

1.9.2.2 Fast Neutron Reactions, (n, p), etc.

Other neutron-induced reactions will normally involve energetic or fast neutrons, where the extra kinetic energy is needed to knock out extra particles. Common reactions are (n, p), (n, α) and (n, 2n), and Figure 1.28 shows these transformations on the *Z* against *N* nuclide chart format. The quantity of radioactivity formed by these reactions is often small because of relatively low fluxes of fast neutrons and small cross sections. However, reactor operators and persons involved in reactor decommissioning will be aware of the significant amounts of activity that can be formed by certain reactions, such as ^{54}Fe(n, p)^{54}Mn, ^{58}Ni(n, p)^{58}Co and ^{27}Al(n, α)^{24}Na. The likelihood of the production of all these radionuclides can again be followed on the nuclide chart.

1.9.2.3 Fission Reactions (n, f)

Figure 1.29 shows that fission products range over masses from about 75 to 165, which correspond to elements Ge to Dy. Most probable nuclides are grouped into two distinct mass regions (the asymmetric mass distribution), as also shown in Figure 1.30.

With ^{235}U(n, f), the distribution peaks twice where $A = 90$–100 and $A = 134$–144. The cumulative fission yield data for each mass number are given on the Karlsruhe Chart as percentage yields on the right-hand edge of the chart.

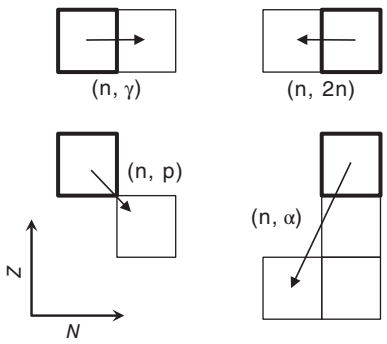


Figure 1.28 Location of products of neutron reactions. Heavy boxes indicate the stable target nuclides.

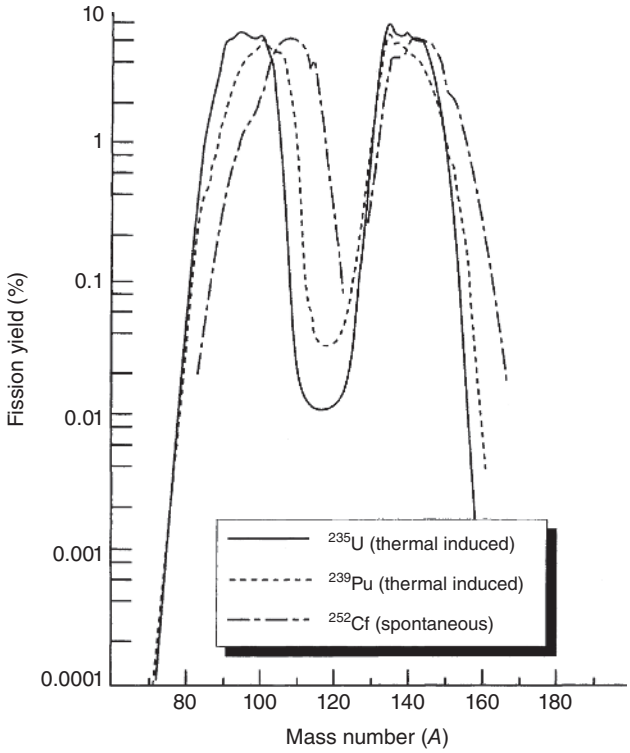
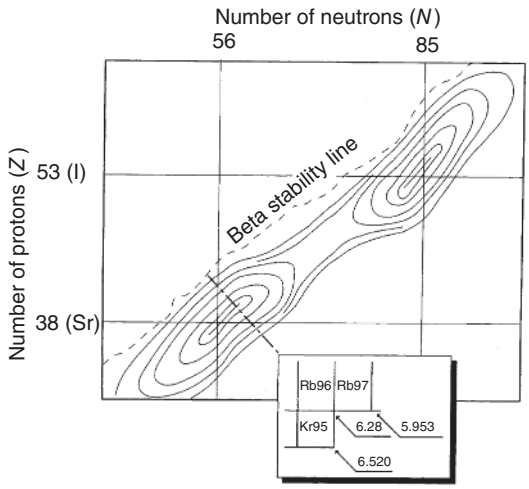


Figure 1.29 Mass yield curves for the thermal neutron fission of ^{235}U and ^{239}Pu, and spontaneous fission of ^{252}Cf.

Figure 1.30 The location of fission products on the Nuclide Chart, indicating regions of high independent yield. The inset shows how data are presented for the cumulative yield of each isobar.



The identification problem appears to be difficult, but bear in mind that

- most fission products fall into narrow bands around the two peaks described earlier, comprising only some 20 isobars;
- all are neutron rich, so that all EC/β^+ nuclides can be ignored;
- a large fraction of possible contenders have short half-lives, and these you may well not need to consider. On the other hand, all fission products are members of isobaric chains and decay will usually result in other activities.

Fission product yields from fast neutron fission show a very similar distribution to Figure 1.29, the main difference being that yields in the minimum between the two peaks increase by a factor of about three.

Information on the history of the 'unknown', in particular, its age or cooling time from the end of irradiation, is most useful. Figure 1.31 shows how the total activity of some elements formed in fission varies with time. These are not all gamma emitters.

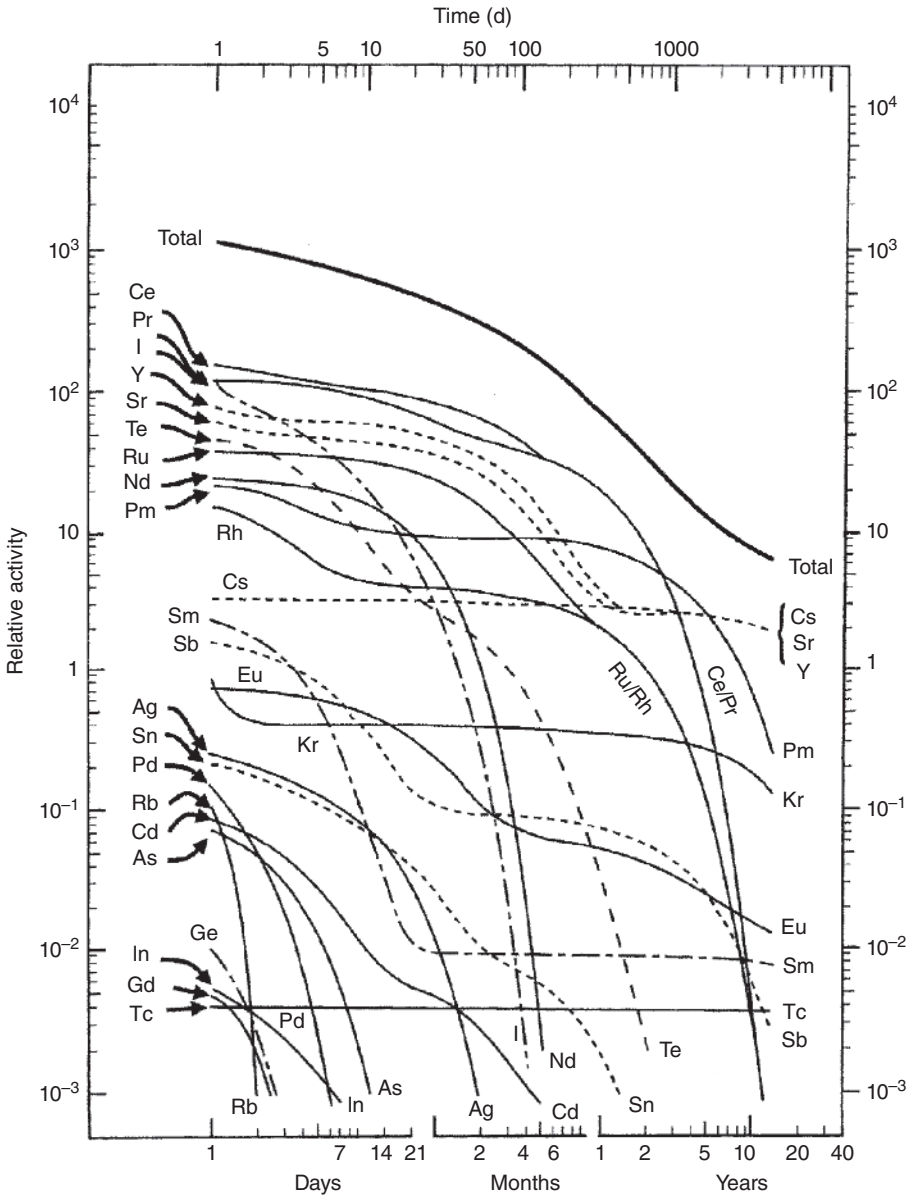


Figure 1.31 Relative radioactivity from fission products as a function of decay time. Data are for thermal neutron fission of ^{235}U, flux of 10^{13} n cm^{-2} s^{-1} and irradiation time of 2 a. Several different nuclides may contribute to the curve for each element (adapted from Choppin and Rydberg (1980)).

Practical Points

- Gamma spectrometry using germanium detectors is the best technique for identifying and quantifying radionuclides. This is due to the very sharply defined and characteristic energies of gamma-rays, which are produced by the great majority of radionuclides.
- *However*, there are a small number of 'pure beta emitters', which do not emit gamma radiation. These cannot be identified by gamma spectrometry. Some are technologically important (^{3}H, ^{14}C, ^{90}Sr).
- Most gamma-rays are a consequence of beta decay. Gamma emission probabilities are not necessarily the same as beta decay probabilities because of internal conversion. This latter can result in X-radiation.
- Gammas and X-rays are usually properties of the daughter nucleus (but not with isomeric transitions). This can lead to identical gamma energies being shown by two isobaric nuclides.
- X-ray energies overlap the low-energy range of gamma-rays. X-ray peak shapes can be different from gamma-ray peak shapes.
- X-ray energies will tell you the element present, but not which isotope. This identification presupposes knowledge of the decay mode: IT → nuclide directly; EC, β^-/IC → daughter.
- Decay schemes give vital information on whether gammas are in 'cascade'. This has great significance in true coincidence summing.
- The Karlsruhe Chart of the Nuclides is a useful tool in helping to identify nuclides, both with regard to classes of nuclide present and for the nuclear data it shows. The data should not necessarily be relied upon for accurate work.

Further Reading

Many of the books referred throughout this book are available on Amazon. However, it may be necessary to search on both Amazon.com and Amazon.co.uk.

• There are a number of general books that cover many of the topics of this chapter:

Keller, C. (1988). *Radiochemistry* (English edition), Ellis Horwood, Chichester, UK.

Ehmann, W.D. and Vance, D.E. (1991). *Radiochemistry and Nuclear Methods of Analysis,* John Wiley & Sons, Inc., New York, NY, USA.

Choppin, G.R., Liljenzin, J.-O., Rydberg, J. and Ekberg C. (2013). *Radiochemistry and Nuclear Chemistry*, 4th Edn, Elsevier. There is also a Kindle version available from Amazon.

A classic authoritative text on the physics of the atom that has run to several later editions:

Evans, R.D. (1955). *The Atomic Nucleus,* McGraw-Hill, New York, NY, USA.

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Browne, E., Firestone R.B., Baglin, C.M. and Chu, S.Y.F. (1998). *Table of Isotopes,* John Wiley & Sons, Inc., New York, NY, USA. See *http://www.wiley.com/toi*.

Fundamental Physical Constants — Extensive Listing. *http://physics.nist.gov/constants*.

• Charts of the nuclides can be found online at:

An interactive online version from the US NNDC (*https://www.nndc.bnl.gov/nudat3/*) containing an overwhelming amount of information.

The Karlsruhe Chart of the Nuclides, in various formats, can be purchased on-line at: *https://nuklid.shop/*.

Printed versions can be found on Amazon as:

Magill, J. and Galey, J. (2004). *Radioactivity, Radionuclides, Radiation (with the Fold-out Karlsruhe Chart of the Nuclides) (Hardcover)*, Springer-Verlag, Berlin, Germany.

The *General Electric* printed version (USA) can also be found on Amazon.com.

In fact, searching Amazon using '*Radioactivity, Radionuclides, Radiation*' will provide a complete list of useful books for anyone starting out in this field.

• For mathematics relating to radionuclide parent-daughter relationships:

Bateman, H. (1910). The solution of a system of differential equations occurring in the theory of radioactive transformations, *Proc. Cambridge Philos. Soc.*, **15**, 423-427. This work is available online at *https://www.whoi.edu/fileserver.do?id=130665&pt=2&p=146929*.