1
Radionuclides and their Radiometric Measurement

1.1
Radionuclides

The first radioactive elements – radium and polonium – were discovered by Marie Curie at the end of the nineteenth century. During the first decades of the twentieth century, tens of natural radioactive elements and their various isotopes in the uranium and thorium decay chains were identified. The first artificial radionuclide, ^{30}P, was produced by Frédéric and Irène Joliot-Curie in an accelerator by bombarding aluminum with protons. Today, more than two thousand artificial radionuclides have been produced and identified, especially after the discovery and use of nuclear fission of uranium and plutonium. This book focuses on radionuclides found in the environment and in nuclear waste. This chapter presents an overview of radionuclides, radioactive decay processes, and the radiometric measurement of radionuclides, which are categorized according to their sources and ways of formation; in later chapters they are classified based on their chemical nature and are discussed in more detail. Radionuclides can be primarily categorized into natural and artificial radionuclides.

1.1.1
Natural Radionuclides

In nature there are three types of radionuclides: those belonging to the decay chains of uranium and thorium, single very long-lived radionuclides, and cosmogenic radionuclides.

The decay chains of uranium and thorium start with two isotopes of uranium and one of thorium, ^{235}U, ^{238}U, and ^{232}Th, which were formed at the birth of the Universe some 13.7 billion years ago, and, since they are so long-lived, they have survived in the earth since its birth 4.5 billion years ago. These three primordial radionuclides each initiate a decay chain leading to the stable lead isotopes ^{207}Pb, ^{206}Pb, and ^{208}Pb, respectively. In between, there are altogether 42 radionuclides of 13 elements, of which nine elements, those heavier than bismuth, have no stable isotopes at all. The three decay chains are depicted in Tables 1.1–1.3. The determination of radionuclides

Table 1.1 Uranium decay chain.

Nuclide	Decay mode	Half-life	Decay energy (MeV)	Decay product
^{238}U	α	4.4×10^9 y	4.270	^{234}Th
^{234}Th	β$^-$	24 d	0.273	^{234}Pa
^{234}Pa	β$^-$	6.7 h	2.197	^{234}U
^{234}U	α	245 500 y	4.859	^{230}Th
^{230}Th	α	75 380 y	4.770	^{226}Ra
^{226}Ra	α	1602 y	4.871	^{222}Rn
^{222}Rn	α	3.8 d	5.590	^{218}Po
^{218}Po	α 99.98%	3.1 min	6.874	^{214}Pb
	β$^-$ 0.02%		2.883	^{218}At
^{218}At	α 99.90%	1.5 s	6.874	^{214}Bi
	β$^-$ 0.10%		2.883	^{218}Rn
^{218}Rn	α	35 ms	7.263	^{214}Po
^{214}Pb	β$^-$	27 min	1.024	^{214}Bi
^{214}Bi	β$^-$ 99.98%	20 min	3.272	^{214}Po
	α 0.02%		5.617	^{210}Tl
^{214}Po	α	0.16 ms	7.883	^{210}Pb
^{210}Tl	β$^-$	1.3 min	5.484	^{210}Pb
^{210}Pb	β$^-$	22.3 y	0.064	^{210}Bi
^{210}Bi	β$^-$ 99.99987%	5.0 d	1.426	^{210}Po
	α 0.00013%		5.982	^{206}Tl
^{210}Po	α	138 d	5.407	^{206}Pb
^{206}Tl	β$^-$	4.2 min	1.533	^{206}Pb
^{206}Pb		stable		

Table 1.2 Actinium decay chain.

Nuclide	Decay mode	Half-life	Decay energy (MeV)	Decay product
^{235}U	α	7.1×10^8 y	4.678	^{231}Th
^{231}Th	β$^-$	26 h	0.391	^{231}Pa
^{231}Pa	α	32,760 y	5.150	^{227}Ac
^{227}Ac	β$^-$ 98.62%	22 y	0.045	^{227}Th
	α 1.38%		5.042	^{223}Fr
^{227}Th	α	19 d	6.147	^{223}Ra
^{223}Fr	β$^-$	22 min	1.149	^{223}Ra
^{223}Ra	α	11 d	5.979	^{219}Rn
^{219}Rn	α	4.0 s	6.946	^{215}Po
^{215}Po	α 99.99977%	1.8 ms	7.527	^{211}Pb
	β$^-$ 0.00023%		0.715	^{215}At
^{215}At	α	0.1 ms	8.178	^{211}Bi
^{211}Pb	β$^-$	36 min	1.367	^{211}Bi
^{211}Bi	α 99.724%	2.1 min	6.751	^{207}Tl
	β$^-$ 0.276%		0.575	^{211}Po
^{211}Po	α	516 ms	7.595	^{207}Pb
^{207}Tl	β$^-$	4.8 min	1.418	^{207}Pb
^{207}Pb		stable		

Table 1.3 Thorium decay chain.

Nuclide	Decay mode	Half-life	Decay energy (MeV)	Decay product
^{232}Th	α	1.41×10^{10} y	4.081	^{228}Ra
^{228}Ra	β$^-$	5.8 y	0.046	^{228}Ac
^{228}Ac	β$^-$	6.3 h	2.124	^{228}Th
^{228}Th	α	1.9 y	5.520	^{224}Ra
^{224}Ra	α	3.6 d	5.789	^{220}Rn
^{220}Rn	α	56 s	6.404	^{216}Po
^{216}Po	α	0.15 s	6.906	^{212}Pb
^{212}Pb	β$^-$	10.6 h	0.570	^{212}Bi
^{212}Bi	β$^-$ 64.06%	61 min	2.252	^{212}Po
	α 35.94%		6.208	^{208}Tl
^{212}Po	α	299 ns	8.955	^{208}Pb
^{208}Tl	β$^-$	3.1 min	4.999	^{208}Pb
^{208}Pb		stable		

in the decay chains has been, and still is, a major topic in analytical radiochemistry. They are alpha and beta emitters, most of which do not emit detectable gamma rays, and thus their determination requires radiochemical separations. This book examines the separations of the following radionuclides: U isotopes, ^{231}Pa, Th isotopes, ^{227}Ac, ^{226,228}Ra, ^{222}Rn, ^{210}Po, and ^{210}Pb.

In addition to ^{235}U, ^{238}U, and ^{232}Th, there are several single very long-lived primordial radionuclides (Table 1.4) which were formed in the same cosmic processes as those that formed uranium and thorium. The most important of these, with respect to the radiation dose to humans, is ^{40}K. However, as this emits readily detectable gamma rays and does not require radiochemical separations, neither this nor the others are discussed further in this book.

The third class of natural radionuclides comprises cosmogenic radionuclides, which are formed in the atmosphere in nuclear reactions due to cosmic radiation (Table 1.5). These radionuclides are isotopes of lighter elements, and their half-lives vary greatly. The primary components of cosmic radiation are high-energy alpha particles and protons, which induce nuclear reactions when they impact on the nuclei of the atmospheric atoms. Most of the cosmogenic radionuclides are attached to aerosol particles and are deposited on the ground. Some, however, are gaseous, such as ^{14}C (as carbon dioxide) and ^{39}Ar (a noble gas), and thus stay in the atmosphere. In

Table 1.4 Some single primordial radionuclides.

Nuclide	Isotopic abundance (%)	Decay mode	Half-life (y)
^{40}K	0.0117	β$^-$	1.26×10^9
^{87}Rb	27.83	β$^-$	4.88×10^{10}
^{123}Te	0.905	EC	1.3×10^{13}
^{144}Nd	23.80	α	2.1×10^{15}
^{174}Hf	0.162	α	2×10^{15}

Table 1.5 Some important cosmogenic radionuclides.

Nuclide	Half-life (y)	Decay mode	Nuclide	Half-life (y)	Decay mode
^{3}H	12.3	beta	^{7}Be	0.15	EC
^{10}Be	2.5×10^6	beta	^{14}C	5730	beta
^{22}Na	2.62	EC	^{26}Al	7.4×10^5	EC
^{32}Si	710	beta	^{32}P	0.038	beta
^{33}P	0.067	beta	^{35}S	0.24	beta
^{36}Cl	3.1×10^5	beta/EC	^{39}Ar	269	beta
^{41}Ca	3.8×10^6	EC	^{129}I	1.57×10^7	beta

primary nuclear reactions, neutrons are also produced, and these induce further nuclear reactions. Two important radionuclides are produced in these neutron-induced reactions: ^{3}H and ^{14}C (reactions 1.1 and 1.2), whose chemistry and radiochemical separations are described in Chapter 13. These radionuclides – tritium and radiocarbon – are generated not only by cosmic radiation but also in other neutron activation processes in nuclear explosions and in matter surrounding nuclear reactors.

$$^{14}\text{N} + \text{n} \rightarrow {}^{14}\text{C} + \text{p} \tag{1.1}$$

$$^{14}\text{N} + \text{n} \rightarrow {}^{12}\text{C} + {}^3\text{H} \tag{1.2}$$

1.1.2
Artificial Radionuclides

Artificial radionuclides form the largest group of radionuclides, comprising more than two thousand nuclides produced since the 1930s. The sources of artificial radionuclides are:

- nuclear weapons production and explosions;
- nuclear energy production;
- radionuclide production by reactors and accelerators.

A wide range of radionuclides are produced in nuclear weapons production, where plutonium is produced by the irradiation of uranium in reactors and in nuclear power reactors. Most are *fission products*, and are generated by the neutron-induced fission of ^{235}U and ^{239}Pu. In nuclear power reactors, they are practically all retained in the nuclear fuel; however, in nuclear explosions they end up in the environment – on the ground in atmospheric explosions or in the geosphere in underground explosions. The spent nuclear fuel from power reactors is stored in disposal repositories deep underground. The radionuclide composition of nuclear explosions and the spent fuel from nuclear power reactors differ somewhat for several reasons. Firstly, the fissions in a reactor are mostly caused by thermal neutrons, while in a bomb fast neutrons are

mostly responsible for the fission events, and this results in differences in the radionuclide composition. Secondly, fission is instantaneous in a bomb, while in a reactor the fuel is irradiated for some years. This allows the ingrowth of some activation products, such as ^{134}Cs, that do not exist in weapons fallout. ^{90}Sr and ^{137}Cs are the most important fission products because of their relatively long half-lives and high fission yields. In addition to these, there is range of long-lived fission products, such as ^{79}Se, ^{99}Tc, ^{126}Sn, ^{129}I, ^{135}Cs, and ^{151}Sm, the radiochemistry of which is discussed in this book.

Along with fission products, activation products are also formed in side reactions accompanying the neutron irradiation. The intensive neutron flux generated in the fission induces activation reactions both in the fuel or weapons material and in the surrounding material. These can be divided into two categories, the first comprising the transuranium elements – a very important class of radionuclides in radiochemistry. These are created by successive neutron activation and beta decay processes starting from ^{238}U or ^{239}Pu (Figure 1.1). Of these, the most important and the most radiotoxic nuclides are ^{237}Np, ^{238,239,240,241}Pu, ^{241,243}Am, and ^{243,244,245}Cm, which are discussed further in this book. In addition to transuranium elements, a new uranium isotope ^{236}U is also formed in neutron activation reactions.

Another activation product group comprises radioisotopes of various lighter elements. In addition to tritium and radiocarbon, a wide range of these activation products are formed in nuclear explosions and especially in nuclear reactors. Elements of the reactor's construction materials, especially the cladding and other metal parts surrounding the nuclear fuel, the steel of the pressure vessel and the shielding concrete structures are activated in the neutron flux from the fuel. Part of these activation products, such as elements released from the steel by corrosion, end up in the nuclear waste disposed of during the use of the reactor. A larger part,
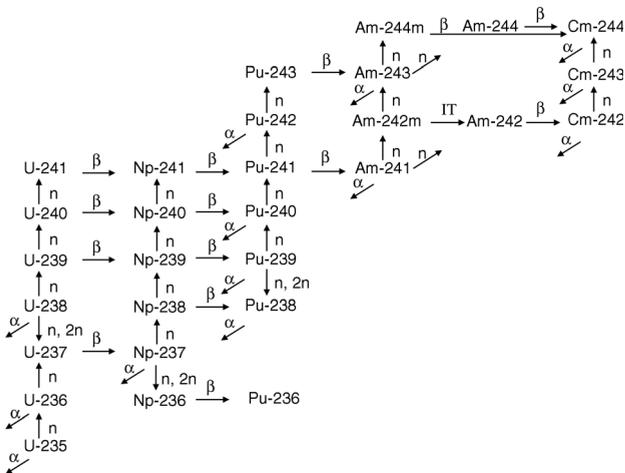
Figure 1.1 Formation of transuranium elements in nuclear fuel and nuclear weapons material (Holm, E., Rioseco, J., and Petterson, H. (1992) Fallout of transuranium elements following the Chernobyl accident. *J. Radioanal. Nucl. Chem. Articles*, **156**, 183).

however, remains in the steel and concrete and ends up in the waste when the reactor is decommissioned. This category has many important radionuclides, such as ^{14}C, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{59}Ni, and ^{63}Ni, which are discussed later in the book. These are all purely beta-decaying radionuclides that require radiochemical separations. In addition to these, there is a range of activation products, such as ^{60}Co, ^{54}Mn, ^{65}Zn, which emit gamma rays and are thus readily detectable and measurable. In addition to the reactor steel and shielding concrete, the spent fuel, its metal cladding, and other metal parts surrounding the fuel and ending up in the final disposal, contain large amounts of the long-lived beta decaying activation products ^{93}Zr, ^{94}Nb, and ^{93}Mo (together with ^{14}C, ^{36}Cl, ^{59}Ni, ^{63}Ni), which are also discussed in this book.

There are also a number of *radionuclides produced by neutron and proton irradiations in reactors and in cyclotrons*. Their properties are later described only if they are used as tracers in radionuclide analysis. An example is a fairly short-lived gamma-emitting strontium isotope, ^{85}Sr, which is used as a tracer in model experiments for studying the behavior of the beta-emitting fission product ^{90}Sr or as a yield-determinant in ^{90}Sr determinations.

1.2
Modes of Radioactive Decay

This book describes the chemistry and analysis of radionuclides – nuclei which are unstable, that is, radioactive. The instability comes from the fact that the mass of the nucleus is either too high or its neutron to proton ratio is inappropriate for stability. By radioactive decay, the nucleus disposes of the mass excess or adjusts the neutron to proton ratio more closely to what is required for stability. The four main radioactive decay modes – fission, alpha decay, beta decay and internal transition – are briefly described below.

1.2.1
Fission

Spontaneous fission is a characteristic radioactive decay mode only for the heaviest elements. In fission, the heavy nucleus divides into two nuclei of lighter elements which are called fission products. Of the naturally occurring isotopes, only ^{238}U decays by spontaneous fission. Only a very minor fraction, 0.005%, of ^{238}U decays by this mode, the rest decaying by alpha mode. Spontaneous fission becomes more prevailing with the heaviest elements, and for some, such as ^{260}No, it is the only way of decay. Considering the production and amounts of fission products, a more important process than spontaneous fission is induced fission: a heavy nucleus absorbs a particle, most usually a neutron, which results in the excitation and further fission of the nucleus (Figure 1.2). There are several fissionable isotopes, of which ^{235}U and ^{239}Pu are the most important from the point of view of the amounts of fission products generated. These two nuclides are not only fissionable but also fissile, that is, they undergo fission in the presence of thermal neutrons, which enables their use as
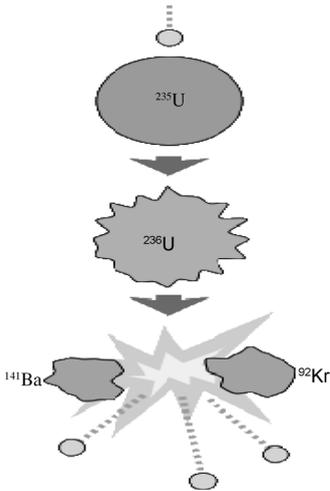
Figure 1.2 An example of a neutron-induced fission of ^{235}U. The reaction is ^{235}U + n → ^{236}U → ^{141}Ba + ^{92}Kr + 3n (http://en.wikipedia.org/wiki/Nuclear_fission).

nuclear fuel in nuclear reactors. ^{235}U is obtained by isotopic enrichment from natural uranium and ^{239}Pu by the irradiation of ^{238}U in a nuclear reactor and subsequent chemical separation of plutonium from the irradiated uranium.

A large number of fission products are generated in fission processes. Figure 1.3 gives, as an example, the distribution of fission products for ^{235}U from thermal
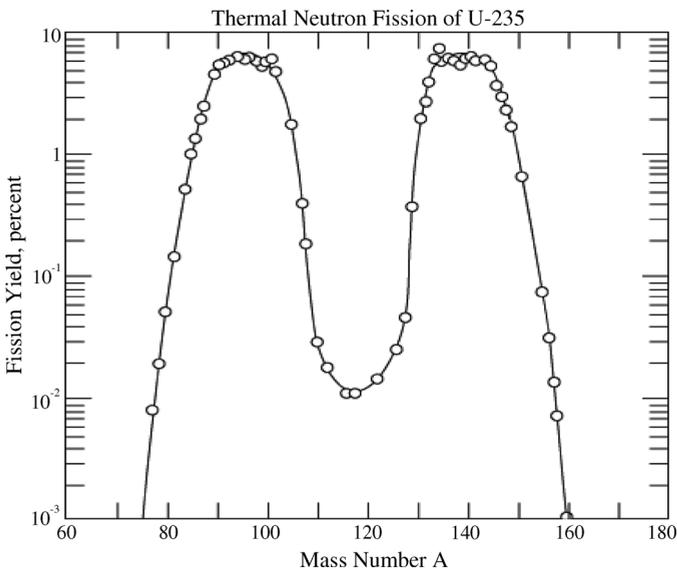
Figure 1.3 Fission yield distribution of ^{235}U as a function of the mass number of the fission product.

neutron-induced fission. As can be seen, the fission is extremely seldom symmetric, that is, the two fission products of one fission event are not of the same mass. Instead, the maxima of fission products are found at the mass numbers 90–100 and at 135–145. At these ranges, the fission yields are between 5% and 7%. This applies to thermal neutron-induced fission; fissions induced by high energy particles become more symmetric with the energy of the bombarding particle.

Most fission products are radioactive since they have an excess of neutrons. In both ^{235}U and ^{239}Pu, the neutron to proton ratio is around 1.6, which is too high for the lighter elements to be stable. For example, for the stable elements in the upper maximum of the fission yield at the mass numbers 135–145, the highest neutron-to-proton ratio is around 1.4, and, through the radioactive decay, by beta minus decay in this case, the nucleus transforms the ratio into an appropriate one. An example of such a decay chain of neutron-rich fission products leading to stable ^{137}Ba is as follows:

$$^{137}\text{Te}\,(t_{1/2} = 3.5\,\text{s}; \text{n/p ratio}\,1.63) \rightarrow {}^{137}\text{I}\,(t_{1/2} = 24.5\,\text{s}; \text{n/p ratio}\,1.58) + \beta^- \rightarrow$$

$$^{137}\text{Xe}\,(t_{1/2} = 3.82\,\text{min}; \text{n/p ratio}\,1.54) + \beta^- \rightarrow$$

$$^{137}\text{Cs}\,(t_{1/2} = 30\,\text{y}; \text{n/p ratio}\,1.49) + \beta^- \rightarrow {}^{137}\text{Ba}\,(\text{stable}; \text{n/p ratio}\,1.45) + \beta^-$$
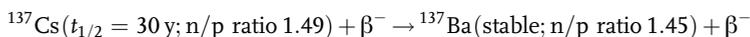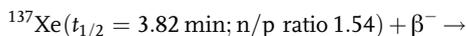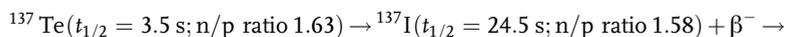
1.2.2
Alpha Decay

Alpha decay is also a typical decay mode for the heavier radionuclides. Most actinide isotopes and radionuclides in the uranium and thorium decay chains decay by this mode. A few exceptions among the radionuclides discussed in this book are ^{210}Pb, ^{228}Ra, and ^{241}Pu, which decay solely by beta emission. ^{227}Ac also decays mostly (98.8%) by beta decay. As can be seen from Tables 1.1–1.3, beta decay is a decay mode competing with alpha decay for many radionuclides in the decay chains. In an alpha decay, the heavy nucleus gets rid of excess mass by emitting a helium nucleus, which is called an alpha particle (α). An example is

$$^{226}\text{Ra} \rightarrow {}^{222}\text{Rn} + {}^{4}\text{He}\,(\alpha) \tag{1.3}$$

where ^{226}Ra turns into ^{222}Rn by emitting an alpha particle. Thus, in an alpha decay, the atomic number decreases by two units and the mass number by four. The energies of the emitted alpha particles are always high, typically between 4 and 7 MeV. Since the mass of the alpha particle is relatively high, the daughter nuclide receives considerable kinetic energy due to recoil. For example, when ^{238}U decays to ^{234}Th by alpha emission, the daughter nuclide ^{234}Th gets 0.074 MeV of the 4.274 MeV decay energy and the alpha particle the rest, 4.202 MeV. Even though the fraction of the recoil energy is only 1.7%, this energy is some ten thousand times higher than that of a chemical bond, and thus recoil results in the breaking of the chemical bond by which the daughter nuclide is bound to the matrix. The transformations from parent nuclides to daughter nuclides take place between defined energy levels corresponding to defined
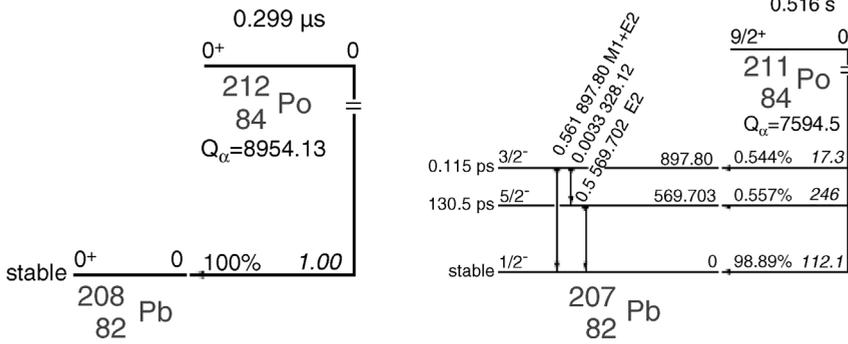
Figure 1.4 Decay schemes of ^{212}Po and ^{211}Po (Firestone, R.B., Shirley, V.S., Chu, S.Y.F., Baglin, C.M., and Zipkin, J. (1996) *Table of Isotopes*, Wiley-Interscience). Upper line: ground state of the parent nuclide; lower line: ground state of the daughter nuclide; intermediate lines: excited states of the daughter nuclide; Q = decay energy (keV); half-life of the parent nuclide at the top; half-lives of the excited states on the left, energies of the excited states in the middle (keV); arrows represent internal transitions and their energies are at their top, percentages are proportions of transitions.

quantum states. Thus, the emitted alpha particles have always the same energy, and peak spectra are obtained when measuring the alpha particles. Often the decay processes lead not only to the ground level of the daughter nuclide but also to its excited energy levels. The alpha particles leading to excited levels have thus lower energies. Since the excited levels, also representing defined quantum states, have defined energy levels, these lower energy alpha particles have defined energies. Because the same energy should be released in each decay process, the rest of the energy, that is, the energy difference between the ground and the excited levels, is emitted as electromagnetic gamma radiation or as conversion electrons. Figure 1.4 gives two schemes of alpha decay processes. The first is for ^{212}Po leading to the ground level of the daughter nuclide only; the other is for ^{211}Po leading also to the excited levels. Later in the text, decay schemes are given for all important radionuclides discussed in this book. The most important information relevant to decay schemes is set out and explained in Figure 1.4.

The fractions of the decay processes going each way, called intensities, have certain probabilities. For example, in the case of ^{211}Po, 98.89% of the decay events go directly to the daughter ground state, while 0.544% go to the upper excitation level and 0.557% to the lower. As the relaxation of the excited levels by gamma emission takes place almost instantaneously after alpha decay in most cases, the excited levels can be considered to decay at the same rate as the parent nuclide. If the intensities of decay processes leading to the gamma ray emissions and the gamma energies are high enough, it is advantageous to measure the activities of the parent nuclides by these gamma emissions since they are often easier to measure than the alpha particles. There are, however, only a few such nuclides. This book discusses in detail those alpha-decaying radionuclides that do not have detectable gamma emissions, since they require radiochemical separations.

1.2.3
Beta Decay

Beta decay is the prevailing decay mode for the lighter radionuclides. The reason for the beta decay is that the nuclide has a too high or too low neutron to proton ratio required for stability. The nucleus adjusts the ratio to a more appropriate one by turning one neutron to a proton or vice versa. The former process takes place with nuclides that have too many neutrons, that is, they are neutron rich; with proton-rich nuclides the latter process takes place. The former process is called beta minus decay (or beta decay) and the latter beta plus decay (or positron decay). In these processes, the nuclei emit particles – a beta particle in the beta minus process and a positron particle in beta plus process. A beta particle (β^-) is physically identical with the electron having the same mass and electrical charge of -1, while the positron (β^+) is an antiparticle of an electron, having the same mass and the same but opposite charge of $+1$. The atomic number (Z) in beta minus decay increases by one unit, while in the beta plus process it decreases by the same amount. In both processes, the mass number (A) remains unchanged. Because of the very small mass of the emitting beta and positron particles, the fraction of the decay energy which the daughter nuclide receives in recoil is very small. The beta particle and the positron do not, however, receive the rest of the decay energy since there is also another particle emitted along with them: a neutrino (ν) with the positron and an antineutrino ($\bar{\nu}$) with beta particle. The complete decay equations are thus:

$$\text{Beta plus decay}: \quad {}_{Z}^{A}M \rightarrow {}_{z+1}^{A}M + \beta^- + \bar{\nu}$$

$$\text{Beta plus decay}: \quad {}_{Z}^{A}M \rightarrow {}_{z-1}^{A}M + \beta^+ + \nu$$
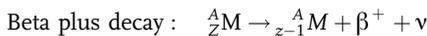
The decay energy is randomly distributed between the particles in the pairs $\beta^-/\bar{\nu}$ and β^+/ν. Since the neutrino and the antineutrino only very weakly interact with the matter, they do not interact with normal detection systems; therefore, when measuring beta-emitting radionuclides, only the beta and positron particles are detected. Since only a fraction of the decay energy goes to these particles, continuous spectra are obtained instead of single peaks corresponding to the decay energy (Figure 1.5). In beta minus decay, the median beta particle energy is about 30% of the maximum energy, while in beta plus decay, the median positron energy is about 40% of the maximum energy. In both spectra the end points of the spectra represent maximum energy, that is, the decay energy.

The positron particles emitted in beta plus decay are not stable. After they have lost their kinetic energy they annihilate by combining themselves with an electron. These two antiparticles are turned into two gamma quanta of 0.511 MeV, an energy corresponding to the mass of an electron. The two gamma rays are emitted in opposite directions.

A competing process for the positron emission in beta plus decay is electron capture (EC). In this process, the proton-rich nuclide, instead of emitting a positron, captures an electron from its atomic electron shell, usually from the innermost K shell and less often from the L shell. This leads to the transformation of a proton into a
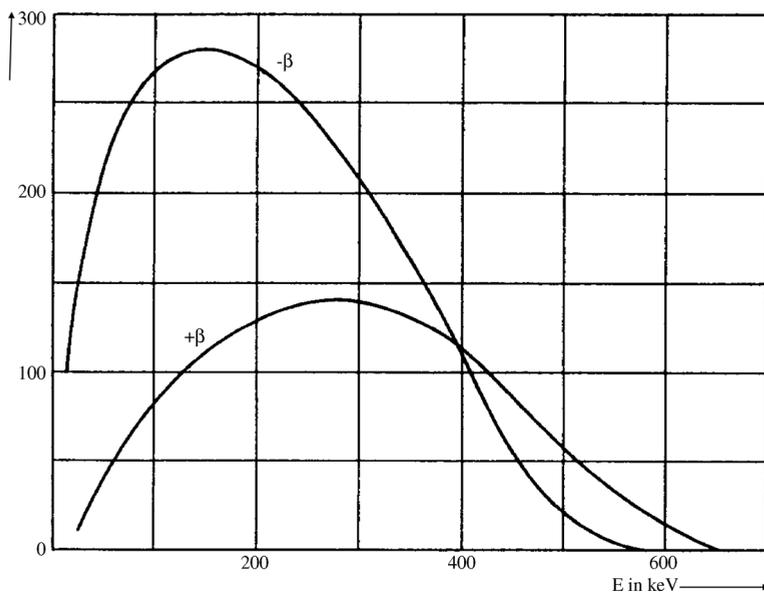
Figure 1.5 Energy spectra of beta and positron particles of ^{64}Cu (Keller, C. (1988) *Radiochemistry*, Ellis Horwood Limited, Chichester).

neutron within the nucleus. In the primary process only neutrinos are emitted from the nucleus. Since these are not detectable, it would seem that it would not be possible to measure the radioactivity of EC-decaying radionuclides. However, EC nuclides can be measured by detecting the X-rays which are formed when the hole in the electron shell is filled by electrons from the upper shells. Though these X-rays are characteristic for the daughter nuclide, they can be used to measure the parent nuclide's activity, since their formation takes place practically simultaneously with the electron capture, and thus their rates are the same. In addition to characteristic X-rays, there are also Auger electrons emitted after electron capture. After the formation of the X-rays, some of these do not leave the atom but transfer their energy to an electron on an upper shell. These Auger electrons, departing from the atom, are monoenergetic and have rather low energies. They can be detected by liquid scintillation counting and thus can be used, in addition to X-rays, to measure activities of the EC nuclides. EC is most typical for the heaviest elements with $Z > 80$, while positron emission is the prevailing decay mode with the lighter elements with $Z < 30$. In between, $30 < Z < 80$, both processes take place in parallel. An example of an EC nuclide dealt with in this book is ^{55}Fe. Its daughter nuclide, ^{55}Mn, emits X-rays with energies of 5.9 keV and 6.5 keV, which are measured for the activity determination of ^{55}Fe. The primary EC process can also lead to an excited state of the daughter nuclide. For example, in the EC decay of ^{125}I, all the primary transitions go to the 35.5-keV excited state of the ^{125}Te daughter nuclide. This excited state will relax by emission of gamma rays, which can be used to measure this and other similar EC nuclides. Thus, there are several options to measure EC nuclides, X-rays, Auger electrons and in some cases
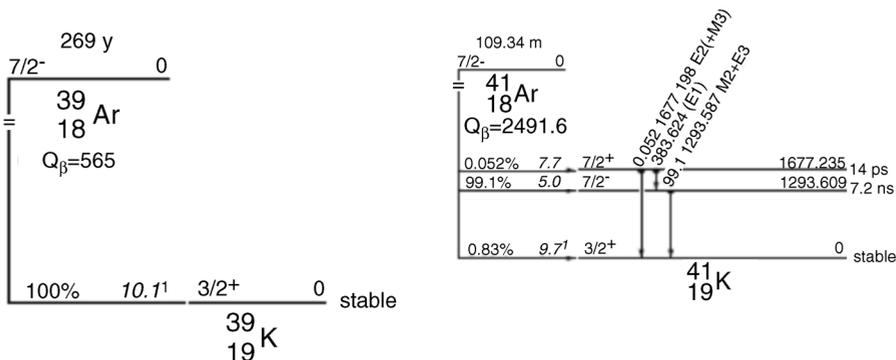
Figure 1.6 Decay schemes of ^{39}Ar and ^{41}Ar (Firestone, R.B., Shirley, V.S., Chu, S.Y.F., Baglin, C.M., and Zipkin, J. (1996) *Table of Isotopes*, Wiley-Interscience).

gamma rays. EC nuclides other than ^{55}Fe discussed in this book are ^{41}Ca, ^{59}Ni, and ^{93}Mo.

As in alpha decay, the transitions in beta decay from the parent to the daughter nuclide often go through excited energy levels of the daughter nuclide; gamma emissions are obtained when these excited levels are de-excited (Figure 1.6). Whenever these gamma emissions have high enough intensities and energies so that they can be readily detected, they are used to measure the activities of beta-emitting radionuclides. This book mostly deals with the pure beta emitters, since they require radiochemical separations. These include ^{3}H, ^{14}C, ^{63}Ni, ^{79}Se, ^{90}Sr, ^{93}Zr, ^{94}Nb, ^{99}Tc, ^{126}Sn, ^{129}I, ^{135}Cs, ^{228}Ra, ^{227}Ac, and ^{241}Pu.

1.2.4
Internal Transition

As mentioned above, alpha and beta decays often go through the excited states of the daughter nuclide. De-excitation of these states takes place in two ways: by emission of gamma rays or by internal conversion. The collective term for these is internal transition. As already described, the excited states represent the defined quantum levels of the daughter nuclide, and therefore the transitions between them have defined energies. Thus, the emitted gamma rays are monoenergetic and the spectrum obtained is a peak spectrum. For example, when ^{41}Ar decays by beta decay (Figure 1.6), only 0.8% of the transitions lead directly to the ground state of the daughter nuclide ^{41}K, the beta decay energy being 2.492 MeV. The remaining 0.05% and 99.1% of the transitions, in turn, lead to the excited states with energy levels of 1.677 MeV and 1.294 MeV, respectively. When these states are de-excited with gamma emissions, the energies of the gamma rays are the same as the difference of the energy of the quantum levels between which the de-excitation takes place. The energies of the beta transitions in Figure 1.6 are the differences between the total beta decay energy (2.492 MeV) and the energies of the excited states (1.677 MeV and 1.294 MeV), that is, 0.815 MeV and 1.198 MeV. Consequently, the gamma rays originating from the de-excitation of these states are 1.677 MeV and 1.294 MeV.
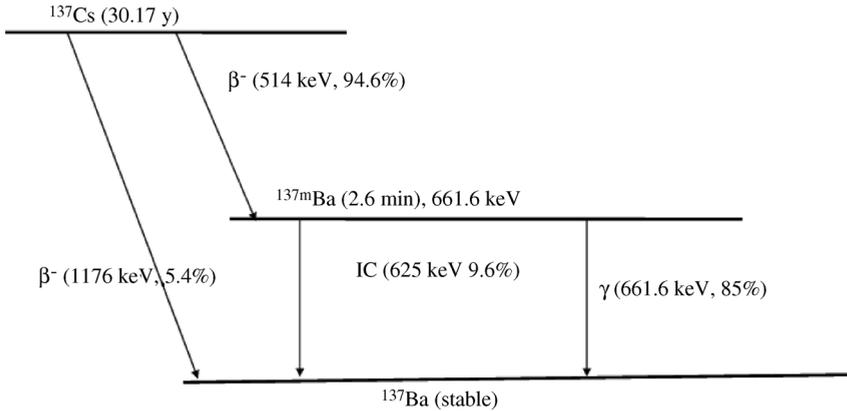
Figure 1.7   Decay scheme of ^{137}Cs.

Many beta-decaying radionuclides emit gamma rays, which are detected to determine the activities of these nuclides.

A competing process for the gamma emission is internal conversion, in which the excited state is not relaxed by the emission of gamma rays; rather, the excitation energy is transferred to an electron at the electron shell of the daughter atom. These electrons are monoenergetic, the energy being the difference between the de-excitation energy and the binding energy of the electron. An example of internal transition is the decay of ^{137}Cs, which partly takes place through the excited state of the daughter nuclide ^{137}Ba (Figure 1.7). Only 5.4% of the transitions go directly to the ground level of ^{137}Ba, the remaining 94.6% going to the 0.662 MeV excited level. This excited state does not solely decay by gamma emissions; 10.2% of the excitations are relaxed by internal conversion and the formation of conversion electrons. The radiations obtained in this decay are:

- 5.4% beta particles with a maximum energy of 1.176 MeV;
- 94.6% beta particles with a maximum energy of 0.514 MeV (1.176–0.662 MeV);
- 85.0% (94.6% × 0.898) gamma rays with an energy of 0.662 MeV;
- 9.6% (94.6% × 0.102) conversion electrons with an energy of 0.625 MeV (0.662–0.037 MeV, where 0.037 MeV is the binding energy of an electron at the K shell of ^{137}Ba). Conversion electrons, the total intensity of which is the above mentioned 9.6%, also have energies of more than 0.625 MeV, since some part of the conversions take place from upper electron shells with lower binding energies.

In addition, there are also X-rays and Auger electrons formed when the electron hole is filled with an electron from an upper shell.

In most cases, the lifetimes of the excited states of the daughter nuclides are very short; they decay simultaneously with the decay of the parent nuclide. In some cases, the lifetimes of the excited states are so long that they can be reasonably easily measured. In these cases, the excited states are considered as independent nuclides and marked with the letter m, meaning metastable. For example, the 0.662 keV excited state of ^{137}Ba has a half-life of 2.55 min and the nuclide is marked as ^{137m}Ba.

The longest half-life of a metastable nuclide has been measured at 241 years for ^{192m}Ir. As already mentioned, the gamma emissions are used to measure the activities of beta-emitting radionuclides since their measurement is often easier. This can only be done if the gamma transitions take place simultaneously with beta decay or when the half-life of the excited state is considerably shorter than the half-life of the beta-decaying nuclide to be measured. An example of the latter is the measurement of ^{137}Cs activity via the 0.662 MeV gamma emissions of ^{137m}Ba. Even though this metastable nuclide has its own half-life, it is very short compared to that of ^{137}Cs (30 years), and thus they both decay at the same rate. When the half-life of the excited state is longer than that of the parent, it cannot be used to measure the activity of the parent since they do not decay at the same rate.

1.3
Detection and Measurement of Radiation

Measurement of radionuclides can be accomplished either by measuring the radiation they emit, that is, radiometrically, or by measuring the number of radioactive atoms by mass spectrometry. The latter methods are described in Chapter 17. In this section, radiometric methods are only summarized. More detailed descriptions of them can be found in a number of textbooks. Radiation measurements are taken in two modes: pulse counting and spectrometry. In pulse counting, the number of pulses is recorded – not their heights. In spectrometry, both the number of pulses and their heights are measured; the pulses are then sorted to the channels of a multichannel analyzer according to their sizes. Pulse counting can only be used for samples containing one single radionuclide or when gross pulse rates are measured. Whenever information on the energy of the pulses is needed, for example in the identification of radionuclides from a mixture, spectrometry is used.

1.3.1
Gas Ionization Detectors

Gas ionization detectors are among the oldest methods still in use for radiation detection and measurement. A gas ionization detector is a chamber filled with an ionizable gas, such as Ar, with an electric field applied across the chamber. The detector is typically an argon gas-filled metal tube, where a metal wire in the middle of the tube acts as the anode, while the tube wall is the cathode (Figure 1.8). When an alpha particle, beta particle, or gamma ray hits the filling gas atoms it loses its energy by ionizing the argon atoms to Ar^+ cations. Because of the electric field applied across the chamber, these cations are collected onto the walls while the electrons go to the anode wire. The electrons are directed out of the tube to an external current circuit and are led back to the tube walls, thus neutralizing the argon cations to form argon atoms. Thus, for each ionization event in the tube, an electric pulse is formed in an external electric circuit. If the pulse height is proportional to initial energy of the particle or gamma/X ray, the system can be used in radiation spectrometry.
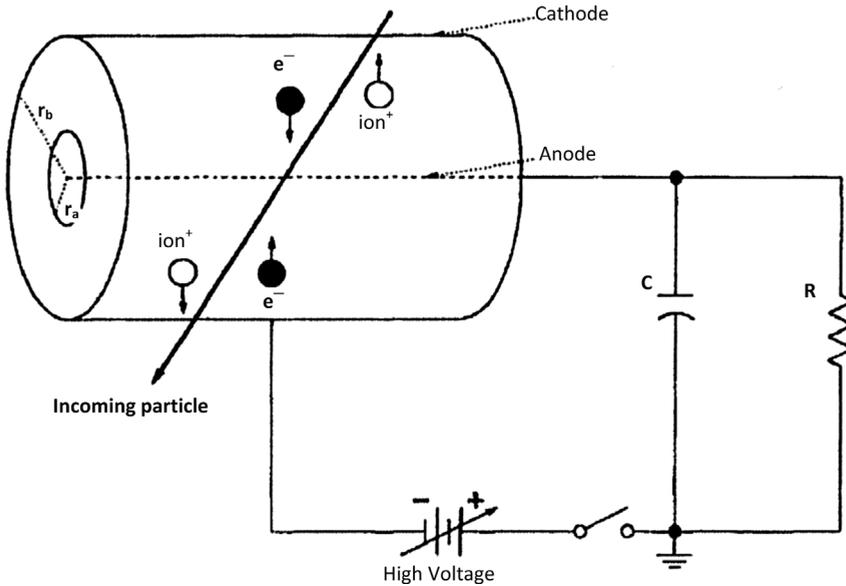
Figure 1.8 Gas ionization detector.

There are basically three types of gas ionization detectors: ionization chamber, proportional counter, and Geiger counter. In the ionization chamber, the electric field applied through the tube is the lowest at about 100–200 volts; the Geiger counter has the highest at some 1000 volts, while in proportional counters the voltage is in an intermediate range. In both ionization chambers and proportional counters, the electric pulse obtained is proportional to the energy lost by a particle or a ray in the filling gas. They can therefore be used to measure energy spectra. The pulse height response in the proportional counter is several orders of magnitude higher than that in the ionization chamber. In the ionization chamber, only ions and electrons in the primary ionization events are recorded, while in the proportional counter the number of ions and electrons is multiplied by a factor of 100–10 000 depending on the voltage used. Because of the very high voltage applied in Geiger counters, they produce pulses that always have the same size, since the filling gas is ionized to its maximum ionization state no matter what the energy of the particle or ray is. For this reason, they can only be used in pulse counting mode. Compared to the proportional counter, even higher voltage pulses are obtained from Geiger counters due to the high voltage, which results in the ionization multiplication by a factor of 10^7–10^{10}. In addition to radiometric measurements, Geiger counters have a wide application area as radiation dose meters used in radiation protection.

In order to be detected, the radiation must get into the chamber. This is usually no problem for gamma radiation since it penetrates the chamber wall. For beta and alpha counting, the other end of the chamber has a thin window through which the particles go into the gas chamber. For low-energy beta radiation, and especially for

alpha radiation, the window needs to be very thin so that the particles do not become adsorbed on it – at least not to great extent. The thinnest films are obtained from organic polymers such as Mylar films made of polyester. Some samples, including gases, can be placed inside the chamber, thus preventing any adsorption on the window.

Gas ionization detectors, especially proportional and Geiger counters, are most widely used in the measurement of beta radiation, excluding the lowest energy beta emitters. Sources for the activity measurements are prepared similarly to sources for alpha counting with semiconductor detectors (see Chapter 5). The sources must be as thin as possible to avoid the absorption of beta particles in the source itself. Typical ways of preparing the counting sources are electrodeposition on a metal plate and microcoprecipitation on a membrane; the former gives a better result because of the lower mass. Furthermore, the sources are placed as close as possible to the detector to obtain a favorable counting geometry and thus an increase in the counting efficiencies. In optimum conditions, the counting efficiencies can be fairly high: 54% for ^{90}Sr/^{90}Y radiation (maximum beta energies 0.546 MeV and 2.280 MeV), 44% for ^{36}Cl (0.709 MeV) and 24% for ^{14}C (0.156 MeV) obtained with the Risö Low-level Beta Counter.

1.3.2
Liquid Scintillation Counting

Liquid scintillation counting (LSC) is a widely utilized radiometric method especially in the measurement of beta-emitting radionuclides. LSC is based on the radiation-induced light formation and transformation of the light into electric pulses. Radionuclide-containing liquid (or solid in a few special cases) is mixed with a liquid scintillation cocktail consisting of an organic solvent, such as toluene, and a scintillating agent. Typically, the radionuclide-cocktail mixture is in 20-mL polyethylene or glass vials, but smaller vials are also available. Scintillating agents are organic aromatic molecules that emit light photons by fluorescence when their excited states are relaxed. Excitations of the scintillator molecules are created by the transfer of the kinetic energy of beta or alpha particles or electrons via solvent molecules to the scintillator molecules. An example of such a scintillating molecule is 1-phenyl-4-phenyloxazole (PPO), which emits light photons at a wavelength of 357 nm. The light pulses penetrate the vial wall and are detected with a photomultiplier tube (PMT) (Figure 1.9).

In a PMT, first a photocathode, consisting of a photosensitive compound such as Cs_3Sb, releases electrons when hit by light pulses. The electrons are multiplied in the PMT with ten or more dynodes, each multiplying the number of electron by a certain factor. This is accomplished by an electric field of about one thousand volts applied through the tube. At the end of the tube, the electrons have been multiplied by a factor of about one million, and the pulse is now high enough to be analyzed further in the system. The height of the electric pulse obtained is proportional to the initial energy of the emitted particle, and for each keV of the particle/ray energy 5–7 light photons are created. For example, when tritium (E_{max} 18 keV) is measured, an average of 100 light photons per beta particle are created in the cocktail; when radiocarbon (E_{max}
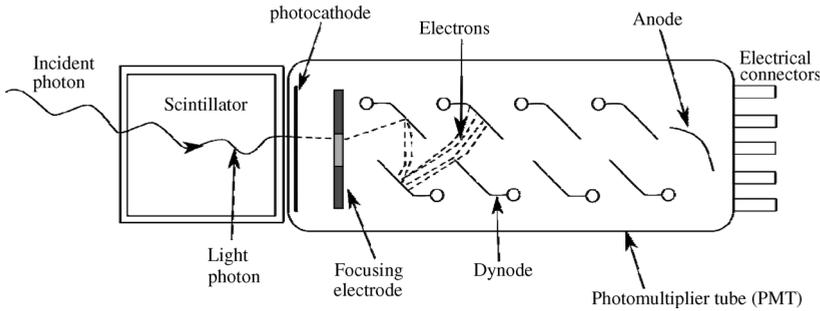
Figure 1.9 Photomultiplier tube coupled to a scintillation detector.

156 keV) is measured, some 300 light photons on average are created. These light pulses release electrons from the photocathode in the same proportion, and since the PMT multiplies these electrons by the same factor, the final electric pulse is proportional to the initial energies of the beta particles.

The liquid scintillation counter has two PMTs facing the sample vial from opposite directions (Figure 1.10) in order to suppress background. The two PMTs are connected in a coincidence mode so that pulses are accepted only if they come from the two multipliers at the same time. Pulses coming from single events on one PMT only, caused by cosmic radiation for example, can thus be avoided. After the coincidence unit, the pulses are summed, further amplified, and turned into digital form in an ADC unit. Finally, the digital pulse goes into a multichannel analyzer which sorts the pulses into different channels based on their pulse height. In this way, an energy spectrum is obtained.

The most challenging tasks in LSC are sample preparation and efficiency calibration. The former is discussed in Chapter 5. Efficiency calibration is needed to take into account the quenching that affects the counting efficiency. Quenching is a decrease in the efficiency of energy transfer from the beta particles, for example, to the scintillator, and a decrease in light photon intensity before they reach the PMTs. The effect of quenching on ^{14}C spectrum can be seen in Figure 1.11 which shows how pulses are lost and the spectrum is shifted to lower channels by quenching.
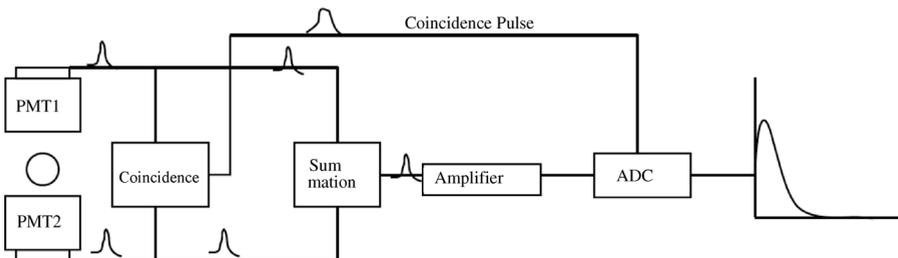

Figure 1.10 Scheme of a liquid scintillation counter (Kessler, M.J. (ed.) (1989). *Liquid Scintillation Analysis, Science and Technology*, Publ. No.169–3052, Perkin-Elmer Life and Analytical Sciences, Boston).
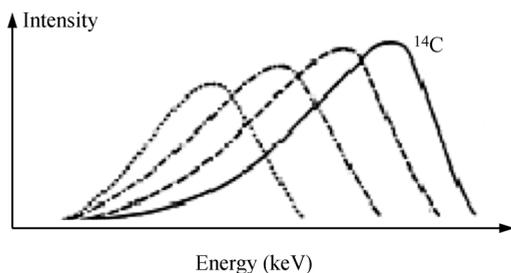
Figure 1.11 The effect of quenching on the LSC spectrum of ^{14}C. The solid line is for an unquenched sample and the broken lines for quenched samples.

There are a number of agents that cause quenching such as acids, alkalis, heavy metals, and alcohols. Since the composition of samples varies, the quenching of different samples also varies. Thus, the counting efficiency must be determined for all samples individually in order to correctly calculate the activity from the measured count rate. This can be done in various ways, of which only a few are described here. The most accurate way to determine the counting efficiency is to use an internal standard. The sample is measured twice: firstly as it is, and secondly after the addition of a known activity amount of the same nuclide. From the difference in count rates, the counting efficiency can be calculated. However, this method is rather laborious because of the double measurement and is thus only seldom used. Other methods include: the sample channels ratio method (SCR), the external standard channels ratio method (ESCR), and the external standard spectral quench parameter (SQP) method. In these methods, a quenching standard curve is created by measuring the counting efficiency for a set of samples containing the same known activity of the studied radionuclide and an increasing amount of quenching agent, such as CCl_4. In the ESCR and SQP methods, the samples are automatically measured twice in the counter: once as it is and then for a short period with an external gamma-emitting source (e.g., ^{226}Ra) positioned below the sample. The gamma rays from the external source cause Compton electrons in the scintillation cocktail, these cause further light emissions in the scintillator and finally electrical pulses in the counter. Increasing the amounts of quenching agent in the cocktail results in a shift to the lower channels of the spectrum created by the Compton electrons. In the SQP method, the end point of the spectrum induced by the external source is recorded and used as a parameter indicating the level of quenching: the higher the quenching the lower is the channel number where the end point is located. From the standard quenching measurements, a curve can now be created where the counting efficiency is plotted against the SQP value (Figure 1.12). Finally, when an unknown sample is measured, the counter produces two primary values: the count rate (imp s^{-1}) and the SQP. From the quenching curve, the counting efficiency is obtained, and this is used to turn the count rate into activity.

Besides beta measurements, LSC can also be effectively used for alpha measurement. The disadvantage of LSC in alpha measurements is its poor resolution. Even at
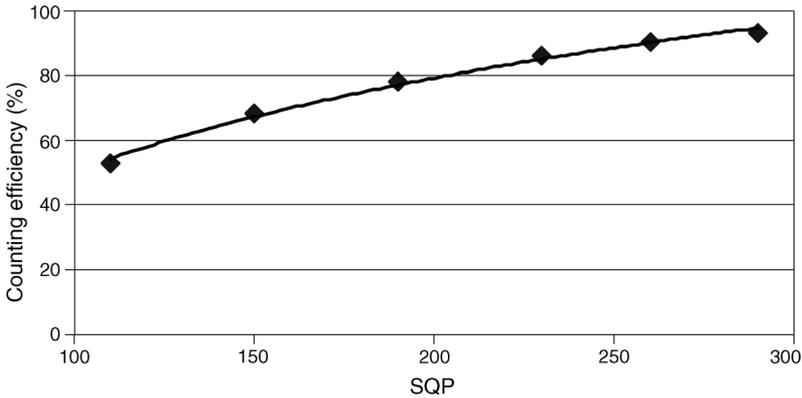
Figure 1.12 Quenching curve for standardization of LSC count rates.

best the resolution is about 200 keV, being ten times higher than in semiconductor measurements. Isotopic information is lost in many cases due to poor resolution. Thus LSC can be mainly used for gross alpha measurements and in cases where high resolution is not needed. Many modern LS counters are able also measure beta and alpha spectra simultaneously from the same sample. This is based on the discrimination of electric pulses caused by beta and alpha radiation due to their different life times: beta pulses are short and alpha pulses last several tens of nanoseconds longer. An example of such a spectrum, where both beta and alpha components are shown, can be seen in Figure 1.13. The alpha/beta discrimination feature facilitates measurement of radionuclide mixtures from natural decay chains in which there are always both beta and alpha emitters present.
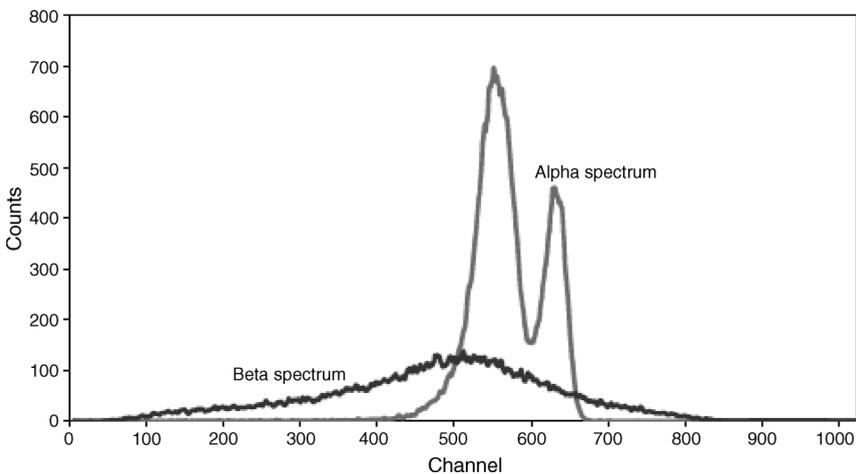

Figure 1.13 LSC spectrum of ground water divided into beta and alpha spectra using alpha/beta pulse discrimination.

The counters used for liquid scintillation counting can also be used for Cherenkov counting. Particles traveling faster than light in water cause the formation of electromagnetic radiation, which is seen as a blue light and called Cherenkov radiation. The PMTs of liquid scintillation counters can detect this light. The minimum energy of a beta particle causing Cherenkov light emissions is 263 keV. Cherenkov counting only becomes practical in activity measurements with nuclides with a mean energy of above 500 keV. The advantage of Cherenkov counting compared to LSC is that no scintillation cocktail is needed and the vial can be completely filled with the sample only. However, the counting efficiency is not as high as with LSC. For example, the counting efficiency for ^{137}Cs with a mean beta energy of 427 keV is 2%. Counting efficiency increases with the beta energy and is already 25% for ^{32}P, which has a mean beta energy of 695 keV.

For measuring EC nuclides, LSC is a good choice, as it can effectively measure Auger electrons created as a secondary radiation in the EC process. The activity of ^{55}Fe, a pure EC nuclide, is typically measured by LSC.

1.3.3
Solid Scintillation Detectors

Solid scintillation detectors are most typically used for gamma counting and spectrometry, and can also be used to measure other types of radiation; however, this is not discussed here. Solid scintillation detectors used in gamma detection typically consist of an inorganic crystal capable of producing light emissions when its excitation states are relaxed. Gamma rays are absorbed in the crystal and cause excitations in the crystal material – most usually NaI doped with a small fraction of thallium ions and thus denoted as NaI(Tl). The purpose of the thallium ions is to act as luminescent centers by which the light emissions take place. There are also stoichiometric compounds used as scintillation detectors, such as $Bi_3Ge_4O_{12}$. The light pulses are detected and amplified with a photomultiplier tube and the pulse is directed through an amplifier and an ADC unit to a multichannel analyzer to create an energy spectrum.

Scintillation crystals have one major advantage: they have very good detection efficiencies. They can be produced as large crystals, which enhances the detection efficiency for penetrative gamma radiation. In addition, they can be manufactured as well-type crystals with a hole in the middle. In these kinds of detectors, the sample can be placed into the hole, which improves the detection geometry and thus the efficiency since the crystal surrounds the sample. The shortcoming of scintillation detectors is their poor energy resolution – some several tens of keVs.

1.3.4
Semiconductor Detectors

Semiconductor detectors are widely used in gamma and alpha spectrometry. They are based on two materials – silicon and germanium – the former being mainly used in alpha detectors and the latter in gamma detectors. In a semiconductor detector, two

semiconducting parts are attached together. One part is an n-type semiconductor with mobile electrons, which is produced by introducing phosphorus atoms with a valence of $+V$ into a framework of germanium or silicon crystals – the valence of both being $+IV$. The other part is a p-type semiconductor with positive holes, achieved by having indium atoms with a valence of $+III$ in the framework of germanium or silicon crystals. When an electric field is applied across the system in a reverse bias mode, a region depleted of holes and electrons is formed at the interface of the n and p type semiconductors. When a gamma ray or alpha particle hits this depleted region, electron-hole pairs are formed in the region and the system becomes conducting. The electric field then produces an electric pulse which can be recorded in the external circuit.

In germanium, the mobility of electrons and positive holes is tens of times higher than that in silicon. The depleted zone is thus much deeper in germanium (several centimeters) than it is in silicon, where it is less than one millimeter. Germanium is thus more suitable as a gamma detector: there is more mass in the depleted zone to absorb the energy of the very penetrating gamma radiation. In addition, germanium has a higher atomic number ($Z = 32$) than silicon ($Z = 14$), so that the formation of photoelectrons, by which the gamma ray energies are principally detected, is more than ten times as probable. Silicon, however, is more suitable for the detection of alpha particles (or low-energy gamma rays and X-rays) since these lose their energy over a very short range and interfering gamma rays penetrate the detector. For these reasons, germanium detectors are made as big as possible for gamma detection, while silicon detectors for alpha measurements are small, only a few mm thick, and have only a very thin depletion layer. In a silicon detector for alpha measurement, the p-type layer facing the alpha source is only tens of nanometers deep to allow the easily absorbable alpha particles to reach the depletion zone.

Germanium semiconductor detectors have superior energy resolution compared to scintillation detectors. The resolution can be as low as 0.5% for 122-keV gamma energy, while it is fifteen times higher for a NaI detector (Figure 1.14). Germanium detectors can thus be used to analyze samples containing even tens of gamma emitting radionuclides at the same time – process which is not possible with a NaI detector. Germanium detectors are therefore valuable tools to identify radionuclides in complex nuclear waste and environmental samples. Special attention is paid in this book to the radionuclides that require radiochemical separations, mostly alpha and beta-emitting radionuclides. For this reason, gamma spectrometry of the samples without radiochemical separations is not discussed in detail.

In conventional gamma detectors, the actual germanium detector is shielded with an aluminum cover which absorbs low-energy gamma rays and often makes their detection impossible. To detect low-energy gamma rays, new detectors have been developed with very thin windows that allow these rays to reach the germanium detector. The windows in these low-energy detectors and broad-energy detectors consist of beryllium or carbon composite. The use of these low-energy gamma detectors enables some radionuclides, such as ^{210}Pb and ^{241}Am, to be detected without radiochemical separation. The detection limit in direct gamma measurement, however, is lower than that in beta or alpha measurement following a
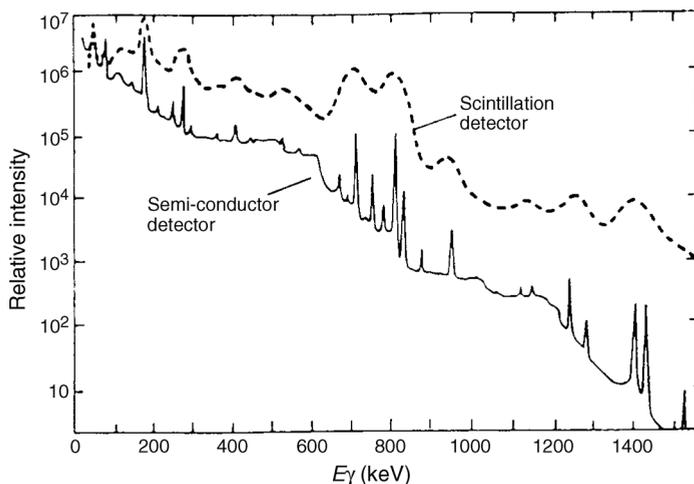
Figure 1.14 Gamma spectrum of [166]Ho measured by a scintillation detector and by a semiconductor detector (Keller, C. (1988) *Radiochemistry*, E. Horwood, Halsted Press, Chichester, New York).

radiochemical separation. Thin-window germanium detectors can also be used to detect X-rays, the measurement of which can be important, as in the case of electron capture radionuclides. As well as thin-window germanium detectors, silicon detectors are used to measure X-rays, but these differ from the silicon detectors used for alpha counting: the X-ray detectors are Li drifted, that is, they contain Li ions as donors.

Unlike gamma spectra, alpha spectra obtained with silicon detectors are, in general, simple to interpret, consisting only of peaks caused by the absorption of alpha particles and only a few pulses due to the background which can be subtracted after separate background measurement; in gamma spectra, however, the background is complicated by the formation of Compton electrons in the detector. Alpha radiation is easily absorbable, and therefore the counting source needs to be prepared with as low mass as possible to prevent self-absorption, which results in a loss of alpha particles and the broadening of the alpha peaks. In addition, the measurements have to be carried out in a vacuum to prevent the absorption of alpha particles in the air between the source and the detector.

1.3.5
Summary of Radiometric Methods

Of the four detector types described above, liquid scintillation counters and gas ionization detectors are the most suitable for beta measurements. The dominant method today in beta measurements is LSC because of its easy sample preparation, high counting efficiency, and high sample throughput rate. The most important advantage of proportional and Geiger counting over LSC is the considerably lower

Table 1.6 Suitable measuring methods for alpha- and beta-emitting radionuclides in the environment and in nuclear waste.

Nuclide	Decay mode	Measurement method
^{3}H	beta	liquid scintillation counting
^{14}C	beta	accelerator mass spectrometry
		liquid scintillation counting, gas ionization detectors
^{36}Cl	beta	accelerator mass spectrometry
		liquid scintillation counting
^{41}Ca	EC	accelerator mass spectrometry
		liquid scintillation counting
^{55}Fe	EC	liquid scintillation counting
^{63}Ni	beta	liquid scintillation counting, gas ionization detector
^{59}Ni	EC	X-ray spectrometry, accelerator mass spectrometry
^{79}Se	beta	liquid scintillation counting, accelerator mass spectrometry
^{85}Kr	beta	gas ionization detectors
^{90}Sr	beta	liquid scintillation counting
		gas ionization detectors
^{93}Zr	beta	liquid scintillation counting, ICP mass spectrometry
^{94}Nb	beta	gamma spectrometry, liquid scintillation counting
^{93}Mo	EC	gamma spectrometry, liquid scintillation counting
^{99}Tc	beta	liquid scintillation counting, ICP mass spectrometry
		gas ionization detectors
^{126}Sn	beta	gamma spectrometry, accelerator mass spectrometry
^{129}I	beta	accelerator mass spectrometry
		neutron activation analysis, liquid scintillation counting
^{135}Cs	beta	ICP mass spectrometry, thermal ionization mass spectrometry
Xe-isotopes	gamma	gamma spectrometry
	beta	gas ionization detectors
^{147}Pm	beta	liquid scintillation counting, ICP mass spectrometry
^{151}Sm	beta	liquid scintillation counting, ICP mass spectrometry
^{210}Pb	beta	liquid scintillation counting
	(gamma)	alpha spectrometry (via ^{210}Po), gamma spectrometry
^{210}Po	alpha	alpha spectrometry
		liquid scintillation counting
^{226}Ra	alpha	alpha spectrometry
		liquid scintillation counting
^{228}Ra	beta	liquid scintillation counting (via ^{228}Ac)
		gas ionization detectors (via ^{228}Ac)
^{227}Ac	beta	alpha spectrometry (via ^{227}Th)
Th-isotopes	alpha, beta	alpha spectrometry
		ICP mass spectrometry, liquid scintillation counting
^{231}Pa	alpha	alpha spectrometry
^{234,235,238}U	alpha	alpha spectrometry
		ICP mass spectrometry, liquid scintillation counting
^{237}Np	alpha	ICP mass spectrometry
^{238,239,240}Pu	alpha	alpha spectrometry
		ICP mass spectrometry (except ^{238}Pu)
^{241}Pu	beta	liquid scintillation counting

(*Continued*)

Table 1.6 (Continued)

Nuclide	Decay mode	Measurement method
^{241}Am	alpha	alpha spectrometry
	(gamma)	liquid scintillation counting, gamma spectrometry
^{242}Cm	alpha	alpha spectrometry

background, which enables lower activities to be measured. For example, the detection limit of the gas ionization method for ^{99}Tc using a Risø Low Level beta counter is 2 mBq, with a counting efficiency of 37% and a background count rate of 0.15 CPM, while the detection limit of LSC for ^{99}Tc using Quantulus ultra low level LSC is 55 mBq, with a counting efficiency of 85% and a background count rate of 5.5 CPM in ^{99}Tc window. Liquid scintillation counting is also a good method to measure radionuclides decaying by electron capture by measuring Auger electrons. Liquid scintillation counting can be used to measure alpha-emitting radionuclides with almost 100% detection efficiency; however, the resolution is poor compared to that of silicon semiconductor detectors, which are most often utilized in alpha counting. For gamma spectrometry, the best choices are germanium detectors, which have very good energy resolution and enable a number of radionuclides at the same sample to be the measured. Solid scintillation detectors can also be used in gamma spectrometry; however, their energy resolutions are poor compared to germanium detectors, and therefore they are mostly used in a single-channel mode to measure only one radionuclide at a time. For most long-lived radionuclides with a low specific activity, mass spectrometric methods give better detection limits than radiometric methods. Table 1.6 gives a list of the radionuclides dealt with in this book together with suitable measuring methods.