CHAPTER 4

Unstable Nuclei and Radioactive Decay

Contents

4.1. Radioactive decay

Radioactive decay is a spontaneous nuclear transformation that has been shown to be unaffected by pressure, temperature, chemical form, etc (except a few very special cases). This insensitivity to extranuclear conditions allows us to characterize radioactive nuclei by their decay period and their mode and energy of decay without regard to their physical or chemical condition.

The time dependence of radioactive decay is expressed in terms of the *half-life* ($t_{½}$), which is the time required for one-half of the radioactive atoms in a sample to undergo decay. In practice this is the time for the measured radioactive intensity (or simply, *radioactivity* of a sample) to decrease to one-half of its previous value (see Fig. 1.1). Half-lives vary from millions of years to fractions of seconds. While half-lives between a minute and a year are easily determined with fairly simple laboratory techniques, the determination of much shorter half-lives requires elaborate techniques with advanced instrumentation. The shortest half-life measurable today is about 10^{-18} s. Consequently, radioactive decay which occurs with a time period less than 10^{-18} s is considered to be instantaneous. At the other extreme, if the half-life of the radioactive decay exceeds 10^{15} y, the decay usually cannot be observed above the normal signal background present in the detectors. Therefore, nuclides which may have half-lives greater than 10^{15} y are normally considered to be stable to radioactive decay. However, a few unstable nuclides with extremely long half-lives, $\geq 10^{20}$ y, have been identified. It should be realized that 10^{15} y is about 10^{5} times larger than the age of the universe.

Radioactive decay involves a transition from a definite quantum state of the original nuclide to a definite quantum state of the product nuclide. The energy difference between the two quantum levels involved in the transition corresponds to the *decay energy*. This decay energy appears in the form of electromagnetic radiation and as the kinetic energy of the products, see Element and Nuclide Index for decay energies.

The mode of radioactive decay is dependent upon the particular nuclide involved. We have seen in Ch. 1 that radioactive decay can be characterized by α-, β-, and γ-radiation. *Alpha-decay* is the emission of helium nuclei. *Beta-decay* is the creation and emission of either electrons or positrons, or the process of electron capture. *Gamma-decay* is the emission of electromagnetic radiation where the transition occurs between energy levels of the same nucleus. An additional mode of radioactive decay is that of *internal conversion* in which a nucleus loses its energy by interaction of the nuclear field with that of the orbital electrons, causing ionization of an electron instead of γ-ray emission. A mode of radioactive decay which is observed only in the heaviest nuclei is that of *spontaneous fission* in which the nucleus dissociates spontaneously into two roughly equal parts. This fission is accompanied by the emission of electromagnetic radiation and of neutrons. In the last decade also some unusual decay modes have been observed for nuclides very far from the stability line, namely *neutron emission* and *proton emission*. A few very rare decay modes like ^{12}C-emission have also been observed.

In the following, for convenience, we sometimes use an abbreviated form for decay reactions, as illustrated for the ^{238}U decay chain in §1.3:

$$^{238}\mathrm{U}(\alpha)\,^{234}\mathrm{Th}(\beta^-)\,^{234}\mathrm{Pa}(\beta^-)\,^{234}\mathrm{U}(\alpha),\ \text{etc.},$$

or, if half-lives are of importance:

$$^{238}\mathrm{U}(\alpha,\ 4.5\times10^{9}\ \mathrm{y})\,^{234}\mathrm{Th}(\beta^-,\ 24\ \mathrm{d})\,^{234}\mathrm{Pa}(\beta^-,\ 1.1\ \mathrm{min})\,^{234}\mathrm{U}(\alpha,\ 2.5\times10^{5}\ \mathrm{y}),\ \text{etc.}$$

In the following chapter we discuss the energetics of the decay processes based on nuclear binding energy considerations and simple mechanics, then we consider the kinetics of the processes. In Ch. 11, where the internal properties of the nuclei are studied, the

explanations of many of the phenomena discussed in this chapter are presented in terms of simple quantum mechanical rules.

4.2. Conservation laws

In radioactive decay – as well as in other nuclear reactions – a number of *conservation laws* must be fulfilled. Such laws place stringent limitations on the events which may occur.

Consider the reaction

$$X_1 + X_2 \rightarrow X_3 + X_4 \tag{4.1}$$

where X represents any nuclear or elementary particle. In induced nuclear reactions X_1 may be the bombarding particle (e.g. a ^{4}He atom in a beam of α-particles) and X_2 the target atom (e.g. ^{14}N atoms), and X_3 and X_4 the products formed (e.g. ^{1}H and ^{17}O).

Sometimes only one product is formed, sometimes more than two. In radioactive decay several products are formed; reaction (4.1) is then better written $X_1 \rightarrow X_2 + X_3$. For generality, however, we discuss the conservation laws for the case (4.1).

For the general reaction (4.1):

(a) The *total energy* of the system must be constant, i.e.

$$E_1 + E_2 = E_3 + E_4 \tag{4.2}$$

where E includes all energy forms: mass energy (§12.2), kinetic energy, electrostatic energy, etc.

(b) The *linear momentum*

$$p = mv \tag{4.3}$$

must be conserved in the system, and thus

$$p_1 + p_2 = p_3 + p_4 \tag{4.4}$$

The connection between kinetic energy E_{kin} and linear momentum is given by the relation

$$E_{\text{kin}} = p^2/(2m) \tag{4.5}$$

(c) The *total charge* (protons and electrons) of the system must be constant, i.e.

$$Z_1 + Z_2 = Z_3 + Z_4 \tag{4.6}$$

where the charge is in electron units.

(d) The *mass number* (number of nucleons) in the system must be constant, i.e.

$$A_1 + A_2 = A_3 + A_4 \tag{4.7}$$

(e) The *total angular momentum* p_I of the system must be conserved, i.e.

$$(p_I)_1 + (p_I)_2 = (p_I)_3 + (p_I)_4 \tag{4.8}$$

Since there exist two types of angular momentum, one caused by orbital movement of the individual nucleons and the other due to the intrinsic spin of the nucleons (internal angular momentum), a more practical formulation of (4.8) is

$$\Delta I = I_3 + I_4 - I_1 - I_2 \tag{4.9}$$

where I is the (total) *nuclear spin quantum number*. The quantum rule is

$$\Delta I = 0, 1, 2, 3, \ldots \tag{4.10}$$

i.e. the change of nuclear spin in a reaction must have an integral value.

The three first laws are general in classical physics; the last two refer particularly to nuclear reactions. In Ch. 10 and 11 other conservation laws are discussed for nuclear reactions, but these are less important in radioactive decay.

4.3. Alpha decay

4.3.1. *Detection*

Alpha particles cause extensive ionization in matter. If the particles are allowed to pass into a gas, the electrons released by the ionization can be collected on a positive electrode to produce a pulse or current. *Ionization chambers* and *proportional counters* are instruments of this kind, which permit the individual counting of each α-particle emitted by a sample. Alpha particles interacting with matter may also cause molecular excitation, which can result in fluorescence. This fluorescence – or *scintillation* – allowed the first observation of individual nuclear particles. The ionization in semiconductors caused by α-particles is now the most common means of detection, see Ch. 8.

4.3.2. *Decay energy*

Alpha decay is observed for the elements heavier than lead and for a few nuclei as light as the lanthanide elements. It can be written symbolically as

$$^{A}_{Z}\mathrm{X} \rightarrow {}^{A-4}_{Z-2}\mathrm{X} + {}^{4}_{2}\mathrm{He} \tag{4.11}$$

We use X to indicate any element defined by its nuclear charge, Z and Z-2 in this equation. Examples are given in Ch. 1, and can be found e.g. in the natural radioactive decay series, see next chapter.

The decay energy can be calculated from the known atomic masses, because the binding energy released (spontaneous decay processes must be exoergic) corresponds to a

disappearance of mass, cf. eqns. (3.2) and (3.5). This energy is also called the *Q-value of the reaction*

$$Q\,(\text{MeV}) = -931.5\,\Delta M\ \ (\text{u}) \tag{4.12}$$

For α-decay we can define the *Q*-value as

$$Q_\alpha = -931.5\,(M_{Z-2} + M_{\text{He}} - M_Z) \tag{4.13}$$

We always write the products minus the reactants within the parenthesis. A decrease in total mass in α-decay means a release of energy. The minus sign before the constant 931.5 is necessary to make *Q* positive for spontaneous decay.

An example will show the use of this equation. For the decay reaction $^{238}\text{U} \rightarrow {}^{234}\text{Th} + {}^{4}\text{He}$, the mass values for ^{238}U and ^{4}He are in Table 3.1; for ^{234}Th it is 234.043 594. Thus we obtain $Q_\alpha = -931.5\,(234.043\,594 + 4.002\,603 - 238.050\,7785) = 4.274$ MeV.

If the products are formed in their ground states, which is common for α-decay, the total decay energy is partitioned into the kinetic energies of the daughter nucleus (E_{Z-2}) and the helium nucleus (E_α):

$$Q_\alpha = E_{Z-2} + E_\alpha \tag{4.14}$$

Because of conservation of energy (4.2) and momentum (4.4)

$$E_{Z-2} = Q_\alpha\, M_\alpha / M_Z \tag{4.15}$$

and

$$E_\alpha = Q_\alpha\, M_{Z-2} / M_Z \tag{4.16}$$

From these equations we can calculate the kinetic energy of the ^{234}Th daughter to be 0.072 MeV, while that of the α-particle is 4.202 MeV. Because of the large mass difference between the α-emitting nucleus and the helium atom, almost all of the energy is carried away with the α-particle.

Although the kinetic energy of the daughter nucleus is small in comparison with that of the α-particle, it is large (72 000 eV) in comparison with chemical binding energies (< 5 eV). Thus the recoiling daughter easily breaks all chemical bonds by which it is bound to other atoms.

In 1904 it was observed by H. Brooks that measurements on ^{218}Po (RaA), obtained from radon, led to a contamination of the detection chamber by ^{214}Pb (RaB) and ^{214}Bi (RaC). This was explained by Rutherford as being due to daughter recoil in the α-decay of ^{218}Po in the sequence (written symbolically):

$$^{222}\text{Rn}(\alpha,\ 3.8\ \text{d})\ ^{218}\text{Po}(\alpha,\ 3.05\ \text{min})\ ^{214}\text{Pb}(\beta^-,\ 27\ \text{min})\ ^{214}\text{Bi}(\beta^-,\ 20\ \text{min})\ldots$$

This recoil led to ejection of ^{214}Pb into the wall of the instrument. The use of the *recoil* of the daughter to effect its separation was employed by O. Hahn beginning in 1909 and played a central role in elucidating the different natural radioactive decay chains.

The recoil may affect such chemical properties as the solubility or dissolution rate of compounds. For example the dissolution of uranium from uranium rich minerals is considerably higher than one would expect from laboratory solubility data because α and U-atom recoil have moved U-atoms away from their normal sites in the mineral.

Alpha-decay energies are most precisely measured in magnetic spectrometers. From (2.5) and (2.10) it is calculated that

$$E_\alpha = 2\mathbf{e}^2 B^2 r^2 / m_{\mathrm{He}} \tag{4.17}$$

From knowledge of the values of **e**, m_{He}, B, and r, E_α can be calculated. A more common technique is to use *semiconductor detectors* combined with pulse height analyzers ("α-spectrometers", Ch. 8).

4.4. Beta decay

4.4.1. *Detection*

Energetic electrons cause ionization and molecular excitation in matter, although the effect is weaker and more difficult to detect than for α-particles. As a result the effect must be amplified for counting individual β-particles. Ionization is used in *proportional* and *Geiger counters*. Scintillation counting can also be used with various detector systems (Ch. 8).

4.4.2. *The* β*-decay process*

The radioactive decay processes which are designated by the general name of β-decay include electron emission (β^- or $^0_{-1}$e), positron emission (β^+ or $^0_{+1}$e) and electron capture (EC). If we use the β-decay of ^{137}Cs as an example, we can write

$$^{137}\mathrm{Cs} \rightarrow {}^{137\mathrm{m}}\mathrm{Ba} + \beta^-$$

This β-decay must occur between discrete quantum levels of the parent nuclide ^{137}Cs and the daughter nuclide $^{137\mathrm{m}}$Ba.

The quantum levels of nuclei are characterized by several quantum numbers, an important one being the *nuclear spin*. The spin value for the ^{137}Cs ground state level is 7/2, while that of $^{137\mathrm{m}}$Ba is 11/2. The electron emitted is an elementary particle of spin 1/2. In nuclear reactions the nuclear angular momentum must be conserved (4.8), which means that in radioactive decay processes the difference in total spin between reactant and products must be an integral value (4.10). Inspection of our example shows that this conservation of spin rule is violated if the reaction is complete as we have written it. The sum of the spin of the $^{137\mathrm{m}}$Ba and of the electron is 11/2 + 1/2 or 6, while that of the ^{137}Cs is 7/2. Therefore, the change in spin (ΔI) in the process would seem to be 5/2 spin units. Inasmuch as this is

a non-integral value, it violates the rule for conservation of angular momentum. Before accounting for this discrepancy let us consider another aspect of β-decay which seems unusual.

Figure 4.1 shows the β-particle spectrum of ^{137}Cs as obtained by a magnetic spectrometer. The β-particle energy is calculated by the relation

$$E_\beta = \mathbf{e}^2 B^2 r^2 / (2 m_e) \tag{4.18}$$

where m_e is the electron relativistic mass. The spectrum shows the number of β-particles as a function of Br, which is proportional to $\sqrt{E_\beta}$ through (4.18). We observe a continuous distribution of energies. This seems to disagree with our earlier statement that decay occurs by change of one nucleus in a definite energy state to another nucleus also in a definite energy state. The two sharp peaks designated K and L at the high energy end of the spectrum are not related to the beta spectrum itself and are discussed later in the chapter (§4.5).

4.4.3. *The neutrino*

This problem of "wrong" spin change and the continuous "non-quantized" spectrum led W. Pauli to the assumption that β-decay involves emission of still another particle which has been named the *neutrino* and given the symbol ν. The neutrino has a spin value of 1/2, an electric charge of 0, and a mass ≈ 0. It is therefore somewhat similar to the photon, which has neither mass, electric charge nor spin. However, while the photon readily interacts with matter, the neutrino does not. In fact the interaction is so unlikely that a neutrino has a very high probability of passing through the entire earth without reacting.

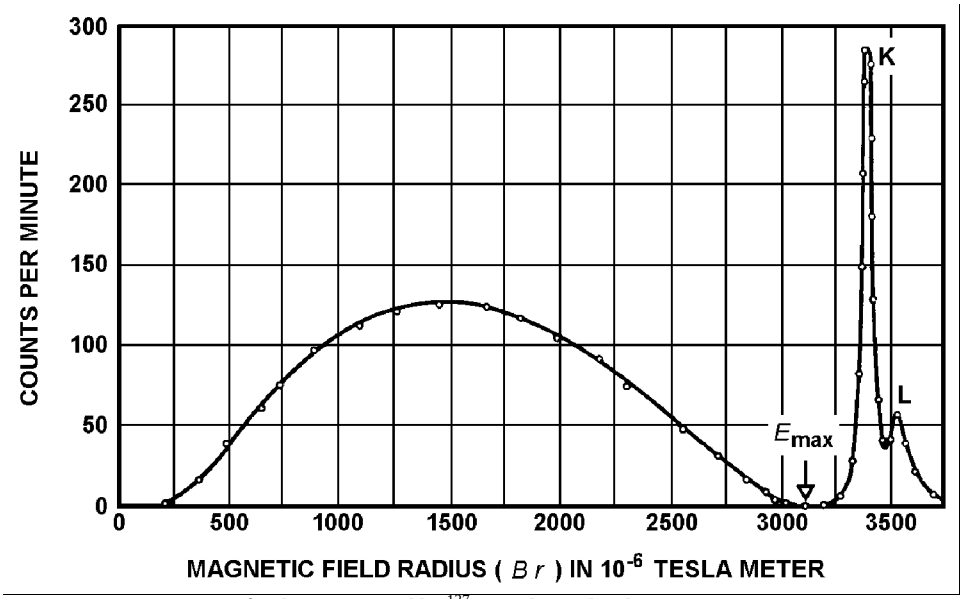


FIG. 4.1. Spectrum for electrons emitted by ^{137}Cs as observed with a magnetic spectrometer. $Br \propto \sqrt{E}$, cf. (4.18). (From W. Gentner, H. Maier-Leibnitz, and W. Bothe.)

The spin attributed to the neutrino allows conservation of angular momentum; in our example, the total spin of the products would be 11/2 + 1/2 + 1/2 or 13/2, and when the spin of ^{137}Cs, 7/2, is subtracted from this the result is 6/2 which is an acceptable integral value. Thus the decay reaction above is incomplete and must be written

$$^{137}\mathrm{Cs} \rightarrow {}^{137\mathrm{m}}\mathrm{Ba} + \beta^- + \bar{\nu}$$

Notice we have replaced ν by $\bar{\nu}$, which is the designation of the *antineutrino*. Beta-decay theory has shown that antineutrinos $\bar{\nu}$ are emitted in electron decay, and "regular" neutrinos ν in positron decay. We can consider the particles identical; cf. §10.4. Because of the extremely low probability of interaction or neutrinos with matter, they are unfortunately often omitted in writing β-decay reactions.

The neutrino theory also explains the energy spectrum in β-decay. However, this necessitates the introduction of another important nuclear concept, that of *relativistic mass* and *rest mass*. In 1901 S. G. Kaufmann showed in experiments that the mass of an electron m seemed to increase when its velocity v approached that of the speed of light **c**. It was found that this increase followed an expression

$$m = m^{\mathrm{o}} (1 - v^2/\mathbf{c}^2)^{-½} \tag{4.19}$$

based on H. Lorentz's studies of the relation between distance, speed of light, and time. m^{o} is the rest mass of the particle (at velocity $v = 0$), while m is referred to as the relativistic mass. This relation is valid for any moving object, macroscopic or microscopic, whether it is a "particle", a "wave-packet" or a space rocket. Figure 4.2 shows $v/\mathbf{c}$ as a function of the kinetic energy of the particle, E_{kin}.

If the parentheses in (4.19) is expanded by means of the binomial theorem of algebra, it approximates to

$$m = m^{\mathrm{o}} + ½\, m^{\mathrm{o}} v^2/\mathbf{c}^2 \tag{4.20}$$

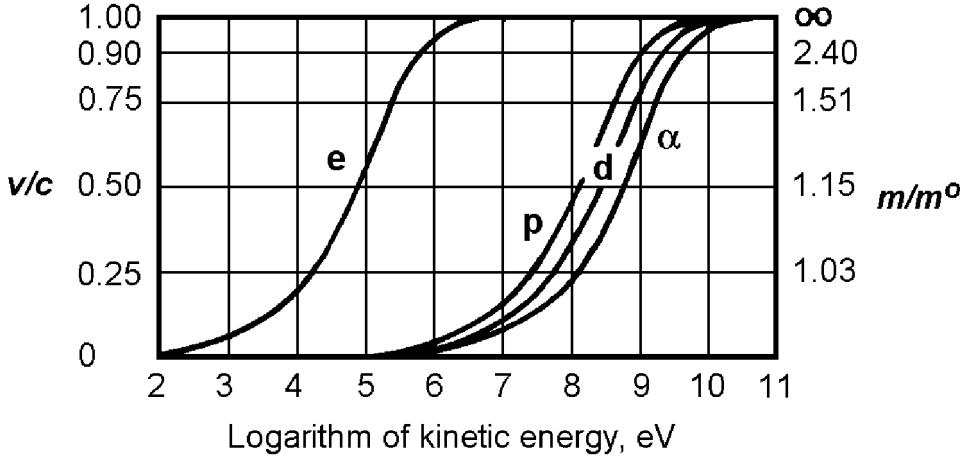


FIG. 4.2. Relativistic masses m for some common nuclear particles, divided by their rest masses m^0, as a function of the kinetic energy of the particle.

The last term is approximately the kinetic energy of the particle (2.5) divided by $\mathbf{c}^2$, and thus

$$m \approx m^o + E_{kin}/\mathbf{c}^2 \tag{4.21}$$

The increase in mass, $\Delta m = m - m^o$, because of the kinetic energy of the particle,

$$E_{kin} = \Delta m\mathbf{c}^2 \tag{4.22}$$

was generalized by A. Einstein in the special theory of relativity, leading (after more detailed calculations) to the well known mass-energy relationship

$$E = m\mathbf{c}^2 \tag{4.23}$$

which we already have applied in the discussion of the nuclear binding energy (3.3).

When a neutrino is ejected from the nucleus it carries away energy of kinetic nature. Thus, according to (4.21) the neutrino has a relativistic mass > 0, and obviously also a momentum $p = mv$. Recoil studies of β-decay have proven this to be true.

In order to correctly apply (4.18) for the calculation of the β-decay energy, the relativistic electron mass must be used; as is seen from Figure 4.2, already at 0.1 MeV, the relativistic mass of the electron is 15% larger than the rest mass $\mathbf{m}_e^o$. (In the following the rest masses of the electron, neutron, etc., will be denoted simply as $\mathbf{m}_e$, $\mathbf{m}_n$, etc.; capital **M** if in universal mass units, u)

The energy released in β-decay is distributed between the neutrino, the electron, and the recoil of the daughter nucleus. This latter will be much smaller than the first two and can be neglected in a first approximation (§4.4.7). Therefore, the total β-decay energy can be considered to be distributed between the neutrino and the electron. For the decay $^{137}Cs \rightarrow {}^{137m}Ba$ it can be shown that the total decay energy Q_β is 0.514 MeV. This is also termed E_{max}. The neutrino energy spectrum is the complement of the β-particle energy spectrum. If the energy of the electron is 0.400 MeV, that of the neutrino is 0.114 MeV. If the electron energy is 0.114 MeV, the neutrino energy is 0.400 MeV.

In β^--decay the average value of the β^--particle energy is approximately $0.3\,E_{max}$. In positron emission, the average energy of the β^+-particle is approximately $0.4\,E_{max}$.

The assumption that the neutrino has a zero rest mass has been questioned by experimentalists and theorists. A number of experiments have established an upper limit of the rest mass as < 10 eV. The implications of a finite rest mass are broad as the nature of the neutrino and the theory of beta decay is involved. On an even grander scale, the expansion of the universe depends on the neutrino mass. If the neutrino rest mass is only a few eV, this might result in sufficiently greater gravitational force that could eventually stop the expansion and contraction will begin, see Ch. 17.

4.4.4. *Double beta decay*

The rather unusual (and very slow) $\beta^-\beta^-$-decay mode is energetically possible for several even-even nuclei, and kinetically possible to observe in those cases where the separating odd-odd nucleus of higher energy prevents normal β^--decay, see Fig. 3.6 V. It has recently been observed for some such cases, e.g.

$$^{82}_{34}\mathrm{Se} \rightarrow {}^{82}_{36}\mathrm{Kr} + 2\beta^- + 2\bar{\nu}$$

The half-life of about 1.7×10^{20} y can be observed because small amounts of the noble gas Kr can be physically isolated from large amounts of ^{82}Se (9% natural abundance), and then measured. Studies of $\beta\beta$-decay are of importance for evaluation of neutrino properties.

4.4.5. β^-*-decay*

This process can be written symbolically as follows:

$$^{A}_{Z}\mathrm{X} \rightarrow {}^{A}_{Z+1}\mathrm{X} + {}^{0}_{-1}\beta + \bar{\nu} \tag{4.24}$$

However, if we take the electrons into account, the neutral parent atom has Z orbital electrons, while the daughter atom, with a nuclear charge of $Z + 1$, must capture an electron from the surroundings, in order to become neutral:

$$^{A}_{Z+1}\mathrm{X}^+ + \mathrm{e}^- \rightarrow {}^{A}_{Z+1}\mathrm{X} \tag{4.25}$$

Moreover, since the negatron emitted provides an electron to the surroundings, the total electron balance remains constant. As a result, in the calculation of the decay energy it is not necessary to include the mass of the emitted β-particle as the use of the mass of the neutral daughter atom includes the extra electron mass. The equation for calculating the Q-value in negatron decay is thus:

$$Q_{\beta^-} = -931.5\,(M_{Z+1} - M_Z) \tag{4.26}$$

As an example we can take the decay of a free neutron in a vacuum; it transforms spontaneously with a half-life of 10.6 min to a proton.

$$^{1}_{0}\mathrm{n} \rightarrow {}^{1}_{1}\mathrm{H} + {}^{0}_{-1}\mathrm{e}^-$$

The Q-value for this reaction is

$$Q_{\beta^-} = -931.5\,(1.007\,825 - 1.008\,665) = 0.782\ \mathrm{MeV}$$

4.4.6. *Positron decay*

Positron decay can be written symbolically as

$$ {}^{A}_{Z}\mathrm{X} \rightarrow {}^{A}_{Z-1}\mathrm{X}^{-} + {}^{0}_{+1}\beta + \nu \rightarrow {}^{A}_{Z-1}\mathrm{X} + {}^{0}_{-1}\mathrm{e}^{-} + {}^{0}_{+1}\beta + \nu \qquad (4.27) $$

Here we must consider the net atomic charges. The daughter nucleus has an atomic number one less than the parent. This means that there will be one extra electron mass associated with the change in atomic number. Moreover, an electron mass must also be included for the positive electron emitted. When ${}^{22}_{11}\mathrm{Na}$ decays to ${}^{22}_{10}\mathrm{Ne}$, there are 11 electrons included in the ^{22}Na atomic mass but only 10 in the ^{22}Ne atomic mass. Consequently, an extra electron mass must be added on the product side in addition to the electron mass associated with the positron particle. The calculation of the *Q*-value must therefore include two electron masses beyond that of the neutral atoms of the parent and daughter

$$ Q_{\beta^+} = -931.5\,(M_{Z-1} + 2\,\mathbf{M}_{\mathrm{e}} - M_Z) \qquad (4.28) $$

Each electron mass has an energy equivalent to 0.511 MeV, since $931.5 \times 0.000\,549 = 0.511$.

Consider the calculation of the *Q*-value for the reaction

$$ {}^{13}_{7}\mathrm{N} \rightarrow {}^{13}_{6}\mathrm{C} + \beta^{+} $$

For this reaction we have

$$ Q_{\beta^+} = -931.5\,(13.003\,355 - 13.005\,739) - 2 \times 0.511 = 1.20 \text{ MeV} $$

4.4.7. *Electron capture*

The EC decay process can be written symbolically

$$ {}^{A}_{Z}\mathrm{X} \overset{\mathrm{EC}}{\rightarrow} {}^{A}_{Z-1}\mathrm{X} + \nu \qquad (4.29) $$

The captured electron comes from one of the inner orbitals of the atom. Depending on the electron shell from which the electron originates, the process is sometimes referred to as K-capture, L-capture, etc. The probability for the capture of an electron from the K-shell is several times greater than that for the capture of an electron from the L-shell, since the wave function of K-electrons is substantially larger at the nucleus than that of L-electrons. Similarly, the probability of capture of electrons in higher order shells decreases with the quantum number of the electron shell.

The calculation of the decay energy in electron capture follows the equation

$$ Q_{\mathrm{EC}} = -931.5\,(M_{Z-1} - M_Z) \qquad (4.30) $$

Note that like the case of the negatron decay, it is not necessary to add or subtract electron masses in the calculation of the *Q*-value in EC. An example of EC is the decay of ^{7}Be to ^{7}Li for which it is possible to calculate that the *Q*-value is 0.861 MeV. This reaction is somewhat exceptional since for neutron deficient nuclei with values of *Z* below 30, positron emission is the normal mode of decay. Electron capture is the predominant mode of decay for neutron deficient nuclei whose atomic number is greater than 80. The two processes compete to differing degrees for the nuclei between atomic numbers 30 and 80. Electron capture is observed through the emission of electrons from secondary reactions occurring in the electron shell because of the elemental change (see §4.9).

4.4.8. *Daughter recoil*

If the β-particle and the neutrino are emitted with the same momentum but in opposite direction, the daughter nucleus experiences no recoil. On the other hand, if they are both emitted in the same direction, or if all the energy is carried away with one of the particles, the daughter experiences maximum recoil. The daughter therefore recoils with kinetic energies from zero up to a maximum value (when the β-particle is emitted with maximum energy). We can therefore write

$$Q_{\beta} = E_{\mathrm{d}} + E_{\max} \tag{4.31}$$

where E_{d} is the recoil energy of the daughter nucleus. From the laws of conservation of energy and momentum, and taking the relativistic mass changes of the electron into account, one finds that the daughter recoil energy is

$$E_{\mathrm{d}} = \mathbf{m}_{\mathrm{e}} E_{\max} / m_{\mathrm{d}} + E_{\max}{}^{2}/(2\, m_{\mathrm{d}}\, \mathbf{c}^{2}) \tag{4.32}$$

The recoil energy is usually ~ 100 eV, which still is sufficient for causing atomic rearrangements in surrounding molecules. In the decay of ^{14}C (to N), $E_{\max}$ is 0.155 MeV, which gives E_{d} = 7 eV. However, by labeling ethane, ^{14}CH $^{14}_{3}$CH , $_{3}$with ^{14}C in both C positions, it was found that $^{14}CH_3NH_2$ was formed in 50% of the cases when one of the ^{14}C atoms in ethane had decayed, although the C≡N bond strength is only 2.1 eV; most of the decays occur with less than the maximum recoil energy, which also can be averaged over the whole molecule. The small recoil also explains why decay reactions like

$$^{127}TeO_3^{2-} \rightarrow {}^{127}IO_3^{-} + \beta^{-}$$

and

$$^{52}MnO_4^{-} \rightarrow {}^{52}CrO_4^{2-} + \beta^{+}$$

are possible, even when E_{d} is tens of electron volts. However, secondary effects tend to cause the chemical bond to break following radioactive decay (see §4.9).

4.5. Gamma emission and internal conversion

The α- and β-decay may leave the daughter nucleus in an excited state. This excitation energy is removed either by γ-ray emission or by a process called *internal conversion*.

The α-emission spectrum of ^{212}Bi is shown in Figure 4.3. It is seen that the majority of the α-particles have an energy of 6.04 MeV, but a considerable fraction (~30%) of the α-particles have higher or lower energies. This can be understood if we assume that the decay of parent ^{212}Bi leads to excited levels of daughter ^{208}Tl. This idea is supported by measurements showing the emission of γ-rays of energies which exactly correspond to the difference between the highest α-energy 6.08 MeV, and the lower ones. For example, an ~0.32 MeV γ accounts for the 5.76 MeV α (6.08 – 5.76 = 0.32). The excited levels of ^{208}Tl are indicated in the insert in Figure 4.3.

Gamma rays produce very low density ionization in gases so they are not usually counted by ionization, proportional, or Geiger counters. However, the fluorescence produced in crystals such as sodium iodide make scintillation counting of γ-rays efficient. Gamma ray spectra can be measured with very high precision using semiconductor detectors (Ch. 8). Figure 4.4 shows such a spectrum for the decay of various excited states of ^{197}Au. The $7/2^+$, etc. symbols are explained in §4.8.

In the great majority of cases the emission of the γ-ray occurs immediately after α- or β-decay, i.e. within $\leq 10^{-12}$ s, but in some instances the nucleus may remain in the higher energy state for a measurable length of time. The longer-lived exited nuclei are called *isomers*. An example is ^{60m}Co, which decays with a half-life of 10.5 min to the ground state of ^{60}Co. The decay is referred to as *isomeric transition*.

The decay energy in γ-emission is distributed between the γ-ray quantum (E_γ) and the kinetic energy of the recoiling product nucleus (E_d). We can therefore write

$$Q_\gamma = E_d + E_\gamma \tag{4.33}$$

The distribution of energy between the γ-ray and the recoiling daughter, according to

$$E_d = E_\gamma^2/(2\, m_d\, c^2) \tag{4.34}$$

shows that $E_d < 0.1\%$ of E_γ. The amount of kinetic energy of the recoiling nuclide is therefore so trivial that it may be neglected when only the γ-ray energy is considered; cf. exercise 4.4.

Gamma rays can interact with the orbital electrons of other atoms, so that the latter are expelled from that atom with a certain kinetic energy (see Ch. 6). A different process, called *internal conversion*, can occur *within* the atom undergoing radioactive decay. Because the wave function of an orbital electron may overlap that of the excited nucleus, the excitation energy of the nucleus can be transferred directly to the orbital electron (without the involvement of a γ-ray), which escapes from the atom with a certain kinetic energy E_e. No γ-ray is emitted in an internal conversion process; it is an alternate mode to γ-ray emission of de-excitation of nuclei.

Internal conversion can be represented symbolically as

$$^{A}_{Z}\mathrm{X}^{m} \rightarrow {}^{A}_{Z}\mathrm{X}^{+} + {}^{0}_{-1}\mathrm{e}^{-} \rightarrow {}^{A}_{Z}\mathrm{X} \tag{4.35}$$

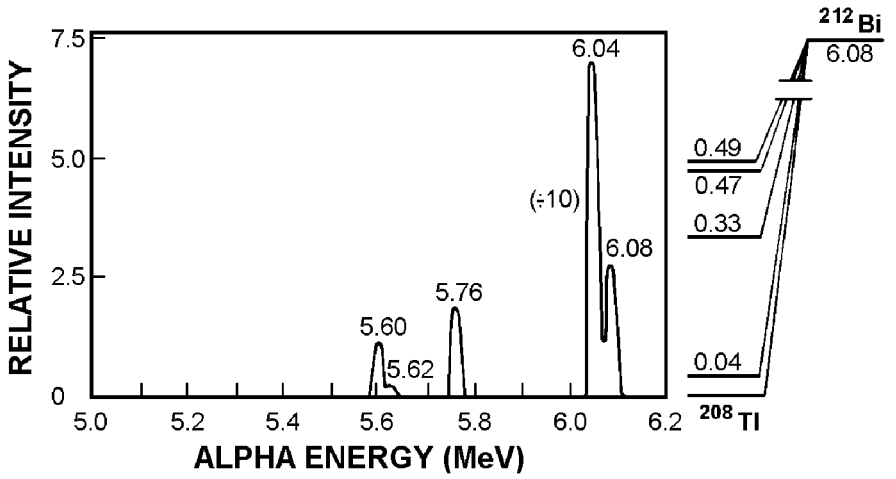


FIG. 4.3. Alpha energy spectrum from $^{212}Bi \rightarrow {}^{208}Tl$. (According to E. B. Paul.)

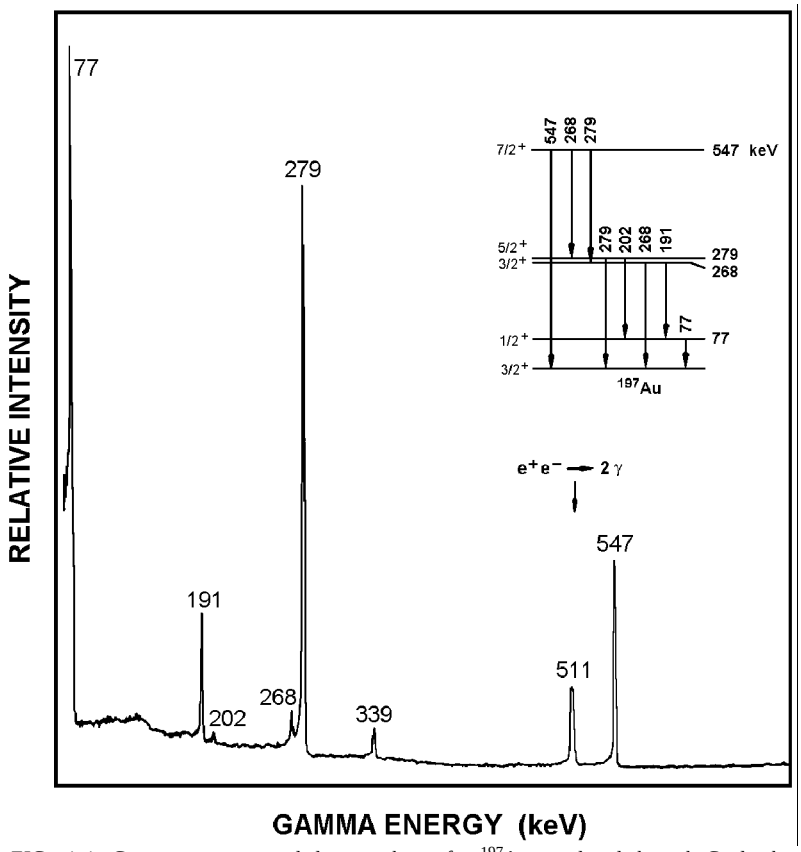


FIG. 4.4. Gamma spectrum and decays scheme for ^{197}Au, produced through Coulomb excitation of gold by a 12 MeV ^{4}He beam. (According to M. G. Bowler.)

where we again have to consider the net atomic charge.

Part of the nuclear excitation energy is required to overcome the binding energy E_{be}, of the electron in its electronic orbital.[1] The remaining excitation energy is distributed between the recoiling daughter nucleus and the ejected electron E_e. The relationship is given by the equation

$$Q_\gamma - E_{be} = E_d + E_e \tag{4.36}$$

The ejected electron, known as the *conversion electron*, normally originates from an inner orbital, since their wave functions have greater overlap with the nucleus. It is to be noted that the conversion electrons are mono-energetic. Inasmuch as the binding energies of the atomic orbitals are different, the values of E_e reflect the differences in the electronic binding energies. In Figure 4.1 two sharp peaks are observed just beyond E_{max}. The first peak, designated as K, is due to conversion electrons originating in the K atomic shell, while the peak labeled L is due to conversion electrons originating in the L atomic shell. Both of these groups of conversion electrons arise from the decay of ^{137m}Ba. Figure 4.5(f) shows schematically the decay process of ^{137}Cs → ^{137}Ba + β^- for the principal decay path; IT is an abbreviation for isomeric transition. The decay of ^{137m}Ba proceeds both by emission of a 0.66 MeV γ-ray and by the competitive process of internal conversion. The ratio between the number of conversion electrons and the number of γ-rays emitted in this competition is called the *conversion coefficient*. The amount of internal conversion is not indicated in simplified decay schemes like Figure 4.5.

If we denote the conversion coefficient as α_i, it is equal to the ratio of K-electrons ejected (which we may denote with I_{eK}) to that of gamma quanta emitted (I_γ):

$$\alpha_K = I_{eK}/I_\gamma \tag{4.37}$$

Usually $\alpha_K < 0.1$. Also $\alpha_K > \alpha_L > \alpha_M$, etc. For ^{137m}Ba the ratio of K-electrons to L-electrons emitted is about 5 while the value of α_K is 0.094.

It can be shown that the energy of the recoiling nucleus (E_d, eqn. (4.36)) is much smaller than the kinetic energy of the ejected electron E_e and may be ignored. The mathematical expression to use is (4.32).

A note on terminology. Consider Figure 4.5(f). Though the γ is emitted from an exited state of ^{137}Ba, in common language we nevertheless talks of "^{137}Cs-γ" (of 0.662 MeV), i.e. as if the γ was emitted from the ^{137}Cs nucleus. The quotation is to be interpreted as "^{137}Cs (decay through β^- followed by) γ(-emission)".

4.6. Spontaneous fission

As the nuclear charge increases to large values, nuclei become more unstable. This is reflected by decreasing half-lives for nuclei heavier than uranium. In 1940 K. Petrzak and

[1] Electron-binding energies are tabulated in standard physics tables.

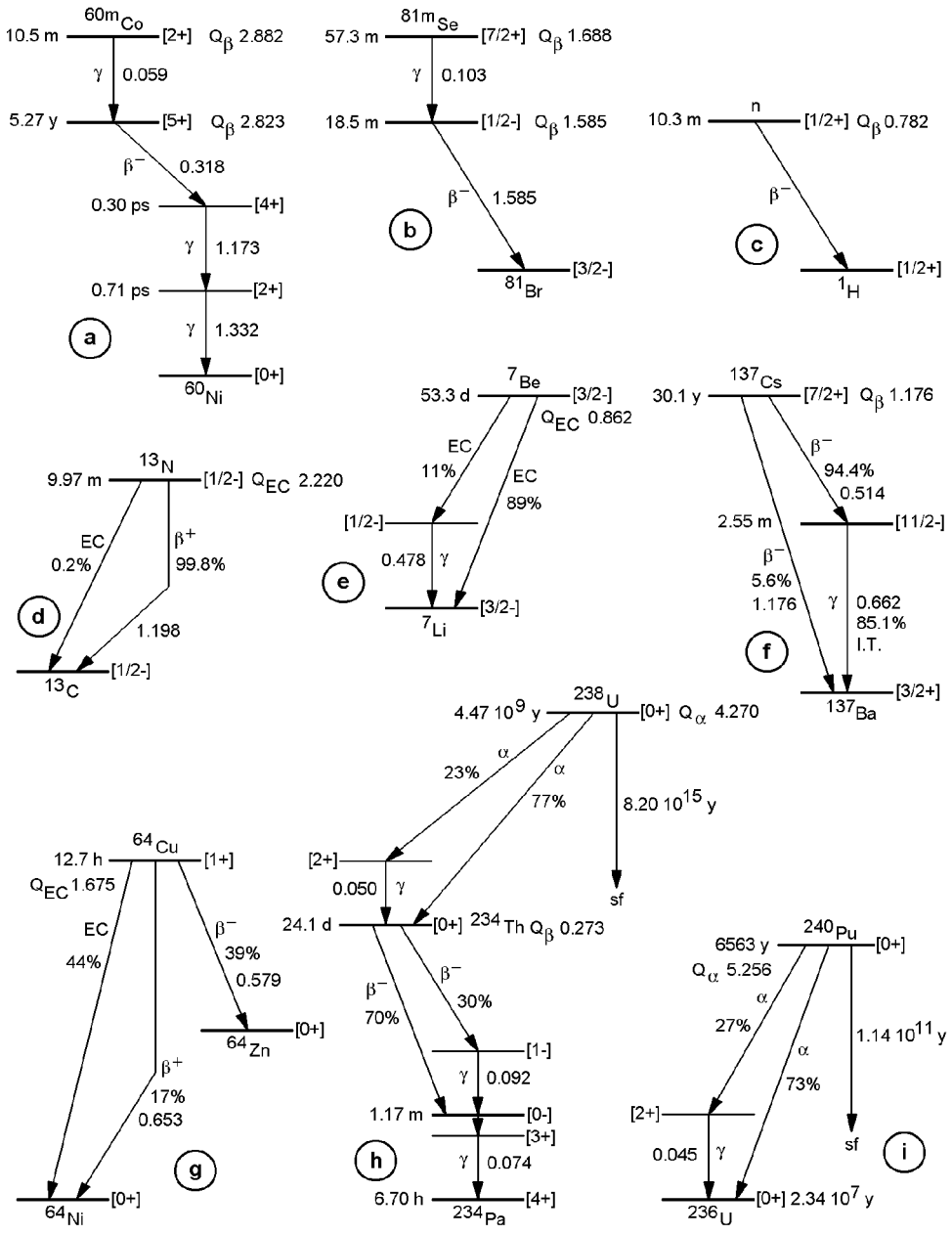


FIG. 4.5. Examples of different decay schemes; energies are in MeV. The schemes are explained in the text.

G. Flerov found that ^{238}U in addition to α-decay also had a competing mode of radioactive decay termed *spontaneous fission*. In this mode two heavy fragments (*fission products*) are formed in addition to some neutrons. The reaction may be written

$$ {}^{A}_{Z}X \rightarrow {}^{A1}_{Z1}X_1 + {}^{A2}_{Z2}X_2 + \nu n \qquad (4.38) $$

where ν is the number of neutrons, usually 2 – 3. The half-life for spontaneous fission of ^{238}U is very long, about 8×10^{15} y. This means that about 70 fissions occur per second in 1 kg of ^{238}U, which can be compared with the simultaneous emission of 45×10^9 α-particles.

With increasing *Z*, spontaneous fission becomes more common; i.e. the half-life for this decay process decreases. For $^{240}_{94}Pu$ it is 1.2×10^{11} y; for $^{244}_{96}Cm$, 1.4×10^7 y; for $^{252}_{98}Cf$, 66 y; and for $^{256}_{100}Fm$, 3×10^{-4} y. In fact, spontaneous fission becomes the dominating decay mode for the heaviest nuclei (see Fig. 3.3).

Spontaneous fission is in some ways similar to fission induced by bombardment with low energy neutrons (§14.7).

4.7. Rare modes of decay

Radioactive decay by *proton emission* is a very seldom observed decay mode for very neutron deficient nuclides because decay by β^+ or EC normally has a very much shorter partial half-life (§4.14). Decay by p^+ has been observed for ^{53m}Co (E_p 1.55 MeV, $t_{½}$ 0.25 s, ~1.5%). However, β^+ decay sometimes leads to a proton-unstable excited state which immediately (< 10^{-12} s) emits a proton. Several β^+ emitters from ^9C to ^{41}Ti with $N = Z - 3$ have β^+ delayed proton emission with half-lives in the range 10^{-3} – 0.5 s. Also radioactive decay by simultaneous emission of two protons has been observed for a few proton rich nuclides, e.g. ^{16}Ne, $t_{½} \sim 10^{-20}$ s.

Among the very neutron rich nuclides, e.g. some fission products, β^- delayed neutron emission is observed. This decay mode is similar in nature to the β^+ delayed p^+ emission. Delayed n-emission is important for the safe operation of nuclear reactors, see Ch. 19.

Decay by emission of particles heavier than α, e.g. ^{12}C, ^{16}O, is energetically possible for some heavy α-emitters and has been observed in a few cases.

4.8. Decay schemes and isotope charts

Information on the mode of decay, the decay energy, and the half-life are included in the *nuclear decay scheme*. A number of simplified decay schemes are shown in Figure 4.5. Figure 3.8 explains a more detailed decay scheme, for $A = 99$.

Using Figure 3.8 as a guide, the decay schemes in Figure 4.5 are easily understood. Figure 4.5(c) shows the β^- decay of the neutron. The decay scheme of ^{137}Cs (Fig. 4.5(f)) differs somewhat from what we would expect from the curve in Figure 4.1. The reason is that in the electron spectrum of Figure 4.1, a small fraction of electrons (8%) emitted with an energy of 1.20 MeV could not be detected because of the insensitivity of the magnetic spectrometer used. It is common for nuclei to decay through different competing reactions, as in this case which involves different β-rays. If the higher energy β-decay had been as

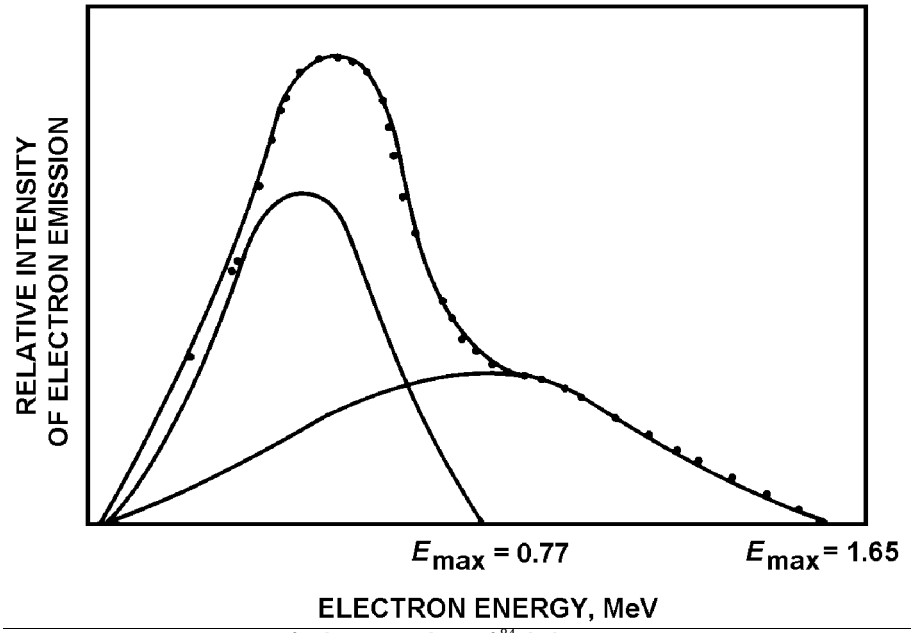


FIG. 4.6. Beta spectrum for the positron decay of ^{84}Rb showing two positron groups, 0.77 MeV (11%) and 1.05 MeV (10%). (^{84}Rb also decays by β^- emission.) (Acc. to S. Eklund)

common as the lower one, we would have observed a mixed β-spectrum, as is indicated in Figure 4.6. Figure 4.5(a) shows the decay of ^{60}Co, and also its isomeric *precursor* ^{60m}Co. The β-decay is immediately followed by a cascade of two γ-rays. ^{64}Cu (Fig. 4.5(g)) decays through negatron (38%) and positron (19%) emission and electron capture (43%); this is referred to as *branched decay*. The vertical line in the angled arrow indicating the positron decay symbolizes the rest mass energy of the two electrons created, i.e. 1.02 MeV. Adding 1.02 MeV to 0.66 MeV gives 1.68 MeV, the *Q*-value for the decay from ^{64}Cu to ^{64}Ni. Figure 4.5(h) is a more complicated decay sequence for ^{238}U(α) ^{234}Th(β^-) ^{234}Pa. In the beginning of this chapter we pointed out that the decay of ^{238}U sometimes results in an excited state of the daughter ^{234}Th (in 23 out of 100 decays), although the excitation energy is comparatively small. Figure 4.5(i) shows how spontaneous fission competes with α-decay in ^{240}Pu. Instead of giving the percentage in the different decay branches, the half-life for that particular mode of decay may be given; conversion between half-lives and percentage is explained in §4.14.

"*Isotope charts*" can be considered as condensed nuclide tables. Figure 3.1 and Appendix C are such charts, though strongly abbreviated. Figure 4.7 shows the beginning of an ordinary chart (i.e. lower left corner of Figure 3.1 and App. C). The legend explains the information provided. Such nuclide charts are very useful for rapid scanning of ways to produce a certain nuclide and to follow its decay modes. Nuclide charts for $_{81}$Tl – $_{92}$U, and for $_{92}$U – $_{109}$Mt are shown in Figures 5.1 and 16.1, respectively.

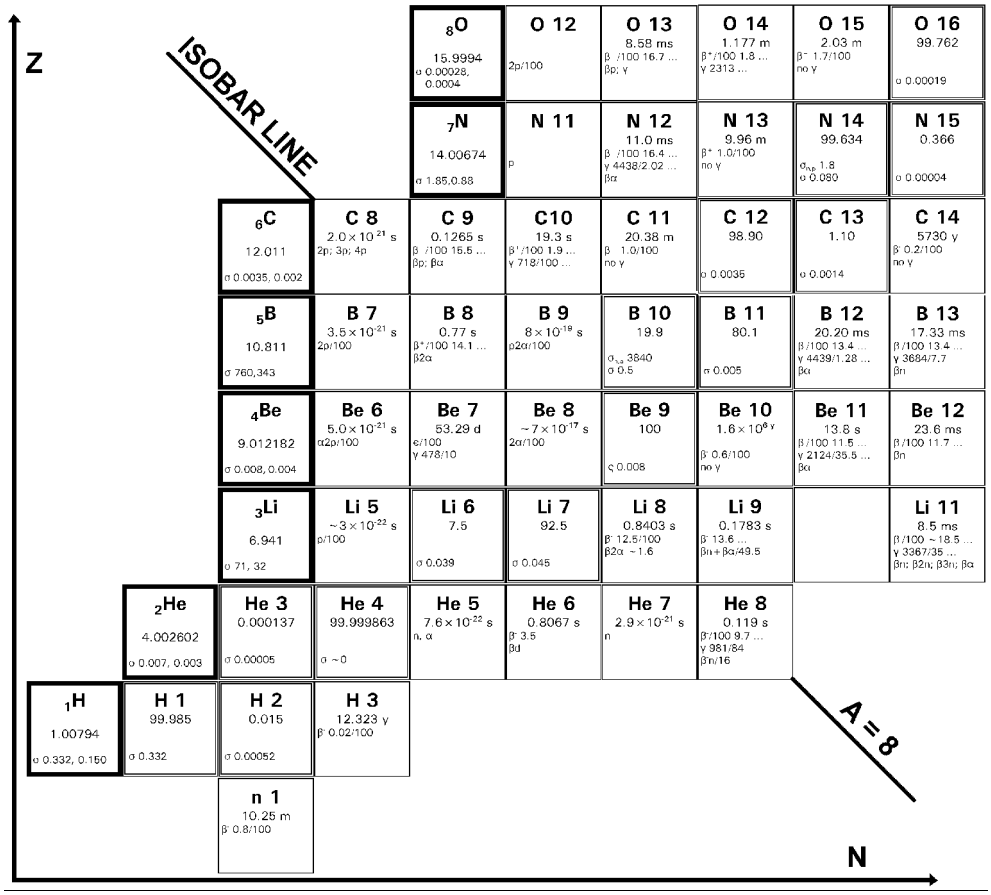


FIG. 4.7. Beginning part of a nuclide chart and schematic nuclear decay and reaction paths.

4.9. Secondary processes in the atom

Once an electron is ejected from an atomic orbital due to internal conversion, electron capture, or some other process involved in radioactive decay, a vacancy is created in the electron shell which can be filled in several ways. Electrons from higher energy orbitals can occupy the vacancy. The difference in the binding energy of the two shells involved in the transition will be emitted from the atom as *X-rays*. This process is called *fluorescent radiation*.

If the difference in the binding energy for the transition is sufficient to exceed the binding energy of electrons in the L- or M-levels, emission of the energy as X-rays is not the predominant mode. Instead an *internal photoelectric* process can occur and the binding energy results in the emission of several low energy electrons which are called *Auger electrons*. Auger electrons are much lower in energy than the electron from the *nuclear internal conversion* process, since the difference in electronic binding energies is in the eV range compared to the energies in the nuclear conversion process which are in the MeV

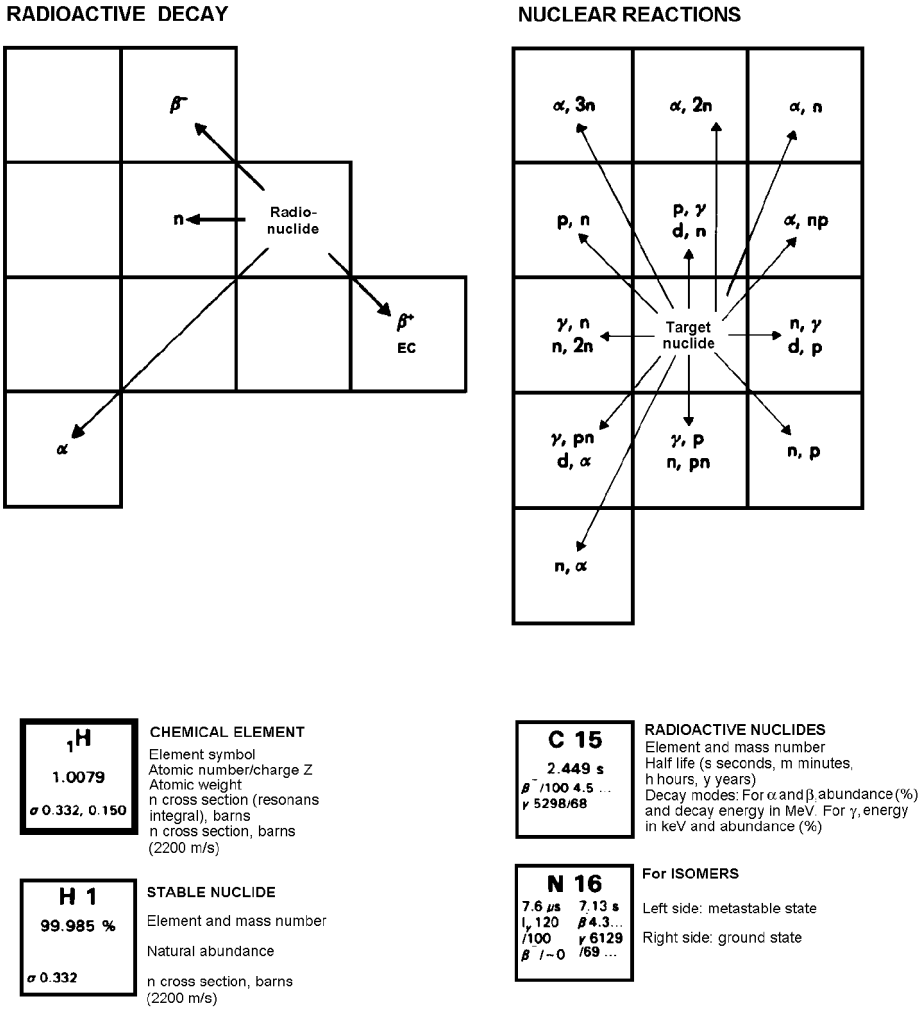


FIG. 4.7. Continued (see §4.20 for sources).

range. The atom may be left in a state of high ionization by Auger emission; positive charges of 10 – 20 have been observed. When such high charges are neutralized, the energy liberated is sufficient to break chemical bonds (cf. §4.4.2).

In isomeric decay the γ-energy is often so small that the daughter recoil is negligible. For example

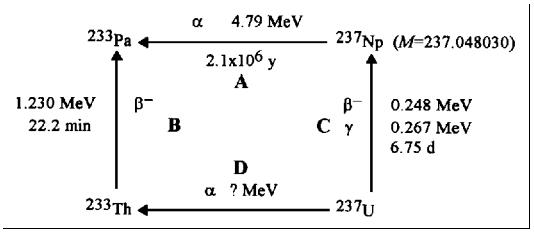
$$^{80m}\mathrm{Br} \xrightarrow[4.4\ \mathrm{h}]{\gamma} {}^{80}\mathrm{Br}$$

occur through the emission of a γ-ray of 0.049 MeV, giving the daughter a recoil energy of only 0.016 eV. Still the decay leads to the emission of ^{80}Br from ethyl bromide, when the parent compound is $C_2H_5{}^{80m}Br$, even though the bond strength is 2.3 eV. This is because the γ is highly converted and as the electron "hole" is filled, Auger emission occurs. Bromine ions from Br^+ to Br^{17+} have been observed through mass spectrometric analyses of the ethyl bromide gas phase.

4.10. Closed decay energy cycles

The masses for many short-lived nuclei are unknown although their decay modes and energies have been determined. From this the nuclear masses may be calculated, and consequently *Q*-values of different unknown decay modes can be obtained. This can be done through the use of closed decay energy cycles.

Suppose we need to know if ^{237}U can decay to ^{233}Th through α-emission. Of course this is a simple calculation if the masses of ^{237}U and ^{233}Th are known, but let us assume they are not. We have data that ^{237}U decays through β-emission (E_{max} 0.248 MeV) followed by γ-decay (E_γ 0.267 MeV). ^{233}Th decays through β-emission (E_{max} 1.230 MeV) directly to ^{233}Pa. ^{237}Np undergoes α-decay to ^{233}Pa with E_α = 4.79 MeV. We may construct a closed cycle including these decay energies



The *Q*-value for branch D is; $Q = -931.5\,(M_{233Th} + M_{He} - M_{237U})$.
For branch A we can calculate (4.16); $Q = E_\alpha\, M_Z / M_{Z-2} = 4.79 \times 237/233 = -931.5\,(M_{233Pa} + M_{He} - M_{237Np})$. By introducing values for M_{He} and M_{237Np} we obtain M_{233Pa} = 233.040 108.
For branch B we calculate; $M_{233Th} = M_{233Pa} + 1.230/931.5 = 233.041\,428$.
For branch C one obtains; $M_{237U} = M_{237Np} + (0.248 + 0.267)/931.5 = 237.048\,581$.

Thus all information is available for calculating branch D. The *Q*-value is found to be 4.23 MeV, and the $E_\alpha = 4.23 \times 233/237$ or 4.16 MeV. Although spontaneous α-decay is energetically possible, it has not been detected. The systematics of α-decay (§4.17) indicates an expected half-life of $> 10^6$ y. Because the β-decay rate is much faster ($t_{½}$ = 6.75 d), too few α's are emitted during the life-time of ^{237}U to be detected.

4.11. Kinetics of simple radioactive decay

Most radioactive isotopes which are found in the elements on earth must have existed for at least as long as the earth. The nonexistence in nature of elements with atomic numbers greater than 92 is explained by the fact that all the isotopes of these elements have life-times considerably shorter than the age of the earth.

Radioactive decay is a random process. Among the atoms in a sample undergoing decay it is not possible to identify which specific atom will be the next to decay. We denote the *decay rate* by A. It is a measure of the number of disintegrations per unit time:

$$A = -\mathrm{d}N/\mathrm{d}t \tag{4.39}$$

The decay rate is proportional to the number of radioactive atoms, N, present: $A \propto N$. If 10^5 atoms show a decay rate of 5 atoms per second then 10^6 atoms show a decay rate of 50 atoms per second. If the number of radioactive nuclei and the number of decays per unit time are sufficiently great to permit a statistical treatment, then

$$-\mathrm{d}N/\mathrm{d}t = \lambda N \tag{4.40a}$$

where λ is the proportionality constant known as the *decay constant*. If the time of observation Δt during which ΔN atoms decay is very small compared to $t_{½}$ (usually $< 1\%$), one may simply write

$$A = \Delta N/\Delta t = \lambda N \tag{4.40b}$$

If the number of nuclei present at some original time $t = 0$ is designated as N_0, (4.40a) upon integration becomes the general equation for simple radioactive decay:

$$N = N_0 \, \mathrm{e}^{-\lambda t} \tag{4.41a}$$

In Figure 4.8 the ratio of the number of nuclei at any time t to the original number at time $t = 0$ (i.e. N/N_0) has been plotted on both a linear (left) and logarithmic (right) scale as a function of t. The linearity of the decay curve in the semi-logarithmic graph illustrates the exponential nature of radioactive decay. Since $A \propto N$, the equation can be rewritten as

$$A = A_0 \, \mathrm{e}^{-\lambda t} \tag{4.41b}$$

Commonly, $\log A$ is plotted as a function of t since it is simpler to determine the disintegration rate than it is to determine the number of radioactive atoms in a sample. Instead of the decay constant λ, the average *lifetime* τ is sometimes used:

$$\tau = 1/\lambda \tag{4.42}$$

Even more common is the use of the *half-life*, $t_{½}$, which is the time needed to reduce the amount of radioactive material by a factor of 2. Thus

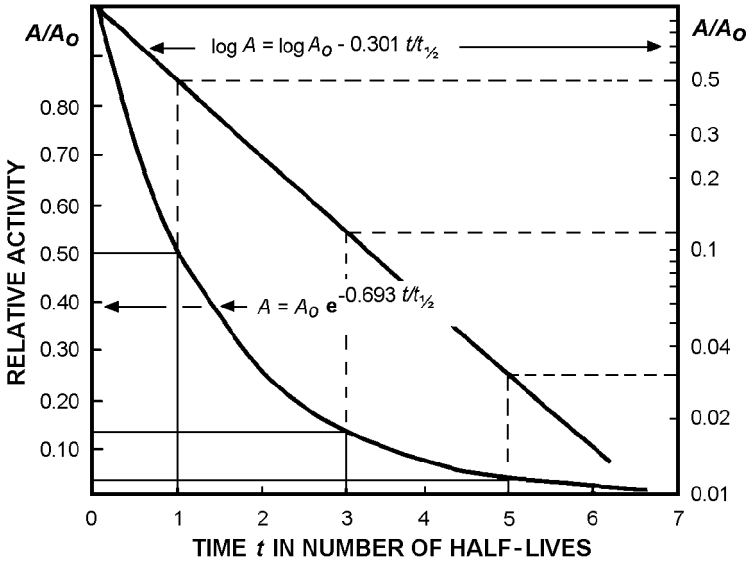


FIG. 4.8. Linear and logarithm plots of simple radioactive decay.

$$A/A_0 = N/N_0 = 1/2 = e^{-\lambda t_{½}}$$

and thus

$$t_{½} = \ln(2)/\lambda = 0.693/\lambda \tag{4.43}$$

$t_{½}$ is about 70% of the average lifetime τ.

The number of radioactive nuclei remaining at any time in a sample which at $t = 0$ had N_0 atoms can be calculated from the equation

$$N = N_0/2^n \tag{4.44}$$

where n is the number of half-lives which have passed. In radioactive work, 10 half-lives ($n = 10$) is usually considered as the useful lifetime for a radioactive species since $N = N_0/2^{10} = 10^{-3} N_0$; i.e. N, and hence A, is 0.001 of the original N_0 and A_0.

The decay rate is usually expressed as disintegrations per second (dps) or disintegrations per minute (dpm). In measuring radioactive decay, it is very rare that every disintegration is counted. However, a proportionality exists for any particular detection system between the absolute disintegration rate A and the observed decay rate:

$$R = \psi A \tag{4.45}$$

where R is the observed decay or *count rate* and ψ the proportionality constant, known as the *counting efficiency*. This counting efficiency depends on many factors including the detector type, the geometry of the counting arrangement, and the type and energy of the

radioactive decay. ψ commonly has a value between 0.01 and 0.5. Equation (4.45) is only valid provided $\Delta t \ll t_{½}$ (in which case $\Delta N \ll N$), where Δt is the time of measurement; this is the normal situation; cf. (4.40b).

Figure 4.9 shows the radioactivity of a ^{32}P sample measured every third day with a GM counter. It is seen that the activity decreases from about 8400 cpm at $t = 0$ to 4200 cpm in 14.3 days, and to about 2100 cpm in 2 × 14.3 days. The uncertainty in the measurements is about the size of the circles, i.e. about ± 110 cpm at $t = 0$ and about ± 65 cpm at 30 days. In plots of this kind, the count rate measured in the absence of the sample (the *background*) must be subtracted from that obtained with the sample present to yield the correct radioactivity for the sample alone. In Figure 4.9 the background is so small (i.e. about 20 cpm) that it has very little influence on the decay curve.

The half-life is such a definitive characteristic of a radioactive species that knowledge of it plus the decay energy is often sufficient to allow identification of a nuclide. A radioactive sample, which exhibits a half-life of 4.5×10^9 y with α-decay energies of 4.8 MeV (77%) and 4.3 MeV (23%), is almost certainly ^{238}U as there is no other nuclide known with this exact set of properties.

With (4.39), (4.43), and (4.45) one obtains

$$R = \psi N \ln(2)/t_{½} \tag{4.46}$$

Knowing the counting efficiency ψ and the number of atoms N, the half-life can be calculated from measurement of R. For example, in a counting arrangement with $\psi = 0.515$ for α-particles, 159 cpm are observed from a ^{232}Th deposit of 1.27 mg (sample weight a). Thus $A = R/\psi = 309$ dpm, $N = a\,N/M = 1.27 \times 10^{-3} \times 6.02 \times 10^{23}/232.0 = 3.295 \times 10^{18}$ ^{232}Th atoms, and $t_{½} = 0.693 \times 3.295 \times 10^{18}/309 = 7.40 \times 10^{15}$ min $= 1.41 \times 10^{10}$ y.

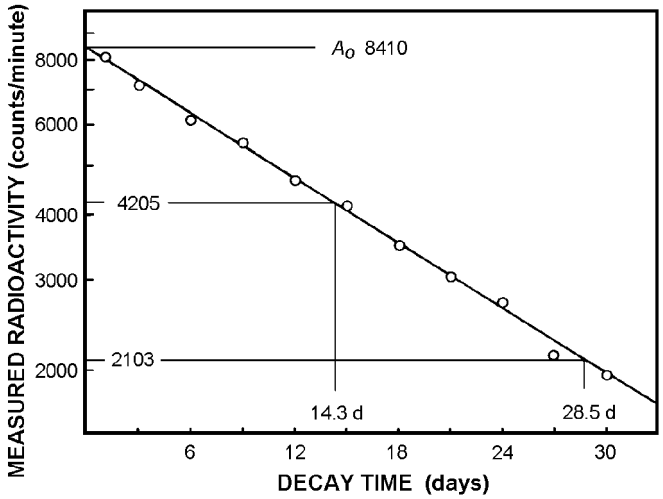


FIG. 4.9. Semilogarithmic plot of the measured decay of ^{32}P using a GM counter with a background of 20 cpm.

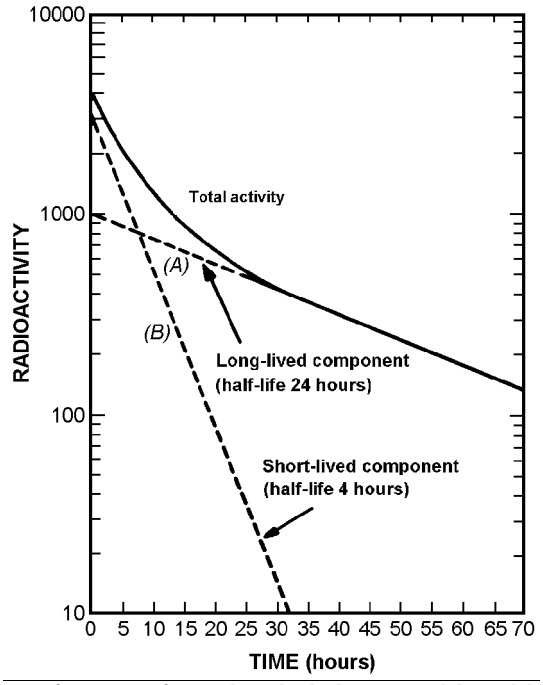


FIG. 4.10. Decay diagram of a mixture of two independently decaying nuclides with half-lives of 4 and 24 h.

It is quite obvious that it would not be possible to determine such a long half-life by following a decay curve like the one in Figure 4.9. Alternately, for short-lived nuclides such as ^{32}P one may use (4.46) with the known half-life and experimentally measured values of ψ and R to determine values of N.

4.12. Mixed decay

A radioactive sample may contain several different radioactive nuclides which are not genetically related. The decay of each nuclide follows the decay equations of the previous section. The detector measures a certain amount of the radioactivity of each species so that

$$R = R' + R'' \tag{4.47a}$$

which with the introduction of (4.41b) and (4.45) gives

$$R = \psi' A_0' e^{-\lambda' t} + \psi'' A_0'' e^{-\lambda'' t} \tag{4.47b}$$

Figure 4.10 shows the composite decay curve for the mixture of ^{71}Zn ($t_{½}$ 3.9 h) and ^{187}W ($t_{½}$ 23.8 h). If the half-lives of the species in the mixture differ sufficiently, as in this case, the decay curve can be resolved into the individual components. The long-lived nuclide (^{187}W, line A) can be observed to have linear decay at times long enough for the

shorter-lived species to have died. The decay line related to this species can be extrapolated back to $t = 0$ and this line subtracted from the observed decay curve. The resulting curve should be the linear decay due to the shorter-lived species in the sample (line B). For more complex mixtures, this process may be repeated until the curve is completely resolved into linear components.

Sometimes it is possible in mixed decay to observe preferentially the decay of one species by proper choice of detection technique. For example, a proportional counter may be used at an operating voltage that allows detection of α-decay ($\psi_\alpha > 0$) but excludes detection of β-decay ($\psi_\beta = 0$). By contrast a typical Geiger counter can be used for β-decay but does not detect α-radiation since the α-particles do not penetrate the window of the Geiger tube. These problems are discussed more extensively in Ch. 8.

4.13. Radioactive decay units

The SI unit for radioactivity is the *Becquerel (Bq)*, and the activity is given in reciprocal seconds, s^{-1}:

$$1 \text{ Becquerel (Bq)} = 1 \text{ (disintegration) } s^{-1} \tag{4.48a}$$

The *measured count rate* (*R*, in (4.45)) is given in *counts per second* (cps) or per minute (cpm); the abbreviations are in German Impulse pro minute (Ipm), and in French coups de minute (c/m).

An earlier unit, still in some use, is the *Curie unit* (abbreviated Ci) defined as:

$$1 \text{ Curie (Ci)} = 3.7 \times 10^{10} \text{ s}^{-1} \text{ (Bq)} \tag{4.48b}$$

The Curie unit was originally defined as the number of decays per unit time and gram of ^{226}Ra, assuming its half-life to be 1580 y.

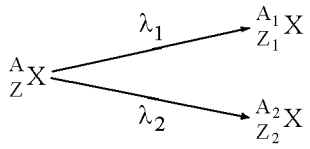
The *specific radioactivity S* is defined as the decay rate *A* per unit amount *w* of an element or compound,

$$S = A/w \tag{4.49}$$

The SI unit of specific radioactivity is Bq kg^{-1}. For practical purposes it is sometimes also defined in dpm g^{-1} or dpm $mole^{-1}$. *Activity concentration* (or "radioactive concentration") is given in Bq m^{-3} or Bq l^{-1}. With the half-life of 1599 ± 4 y the specific activity per gram of ^{226}Ra is 0.988 Ci or 3.65×10^{10} Bq or 2.19×10^{12} dpm. The specific activities of some of the longer-lived naturally occurring radioactive species are: K, 31.3 kBq kg^{-1}; ^{232}Th, 4.05 MBq kg^{-1}; ^{238}U, 12.4 MBq kg^{-1}.

4.14. Branching decay

Several times in this chapter the possibility of competing modes of decay has been noted; see e.g. Figure 1.2. In such competition, termed *branching decay* (see Fig. 4.5(d)), the parent nuclide may decay to two or more different daughter nuclides: e.g.



(4.50)

where for each branching decay a *partial decay constant* can be determined. These constants are related to the total observed decay constant for the parent nuclide as

$$\lambda_{tot} = \lambda_1 + \lambda_2 + \ldots \tag{4.51}$$

Each mode of decay in branching may be treated separately; the decay in an individual branch has a half-life based on the partial decay constant. Since only the total decay constant (the rate with which the mother nuclide, ${}^{A}_{Z}X$ in (4.50), decays) is observable directly, partial decay constants are obtained by multiplying the observed total decay constant by the fraction of parent decay corresponding to that branch. ${}^{64}Cu$ decays 43% by electron capture, 38% by negatron emission, and 19% by positron emission. The observed total decay constant is equal to 0.0541 h^{-1} based on the half-life of 12.8 h. The partial constants are:

$$\lambda_{EC} = 0.43 \times 0.0541 = 0.0233\ h^{-1}$$

$$\lambda_{\beta^-} = 0.38 \times 0.0541 = 0.0206\ h^{-1}$$

$$\lambda_{\beta^+} = 0.19 \times 0.0541 = 0.0103\ h^{-1}$$

These partial decay constants correspond to partial half-lives of 29.7 h for electron capture decay, 33.6 h for β^- decay, and 67.5 h for positron decay.

4.15. Successive radioactive decay

There are many instances where a parent decays to a daughter which itself decays to a third species (i.e. a "grand-daughter"). The chains of radioactive decay in the naturally occurring heavy elements include as many as 10 – 12 successive steps (Fig. 1.2).

$$X_1 \xrightarrow{\lambda_1} X_2 \xrightarrow{\lambda_2} X_3 \xrightarrow{\lambda_3} X_4 \ldots \tag{4.52}$$

The net rate of formation of the daughter atoms X_2 is the difference between the rate of formation of the daughter and her rate of decay, i.e.

$$dN_2/dt = N_1 \lambda_1 - N_2 \lambda_2 \tag{4.53}$$

where N_1 and N_2 are the number of parent and of daughter atoms, and λ_1 and λ_2, the decay constants of the parent and daughter, respectively. The solution of this equation is

$$N_2 = [\lambda_1 /(\lambda_2 - \lambda_1)] N_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t} \tag{4.54}$$

where N_1^0 and N_2^0 are the amounts of parent and daughter respectively at time $t = 0$. The first term in this equation tells us how the number of daughter nuclei vary with time as a consequence of the formation and subsequent decay of the daughter nuclei, while the second term accounts for the decay of those daughter nuclei that were present at $t = 0$.

Let us illustrate this relationship by an example among the naturally occurring radioactive decay series. In an old uranium mineral all the products in the decay chain can be detected (see Fig. 1.2). Suppose now that we use a chemical separation to isolate two samples, one containing only uranium and one containing only thorium (relation (1.3) and Fig. 1.1). At the time of separation, which we designate as $t = 0$, there are N_1^0 atoms of ^{238}U and N_2^0 atoms of ^{234}Th. In the thorium fraction, which is free from uranium, $N_1^0 = 0$ and, therefore, the thorium atoms decay according to the last term in (4.54). This sample gives a simple exponential decay curve with a half-life of 24.1 d, as shown by the precipitate curve in Figure 1.1. The uranium fraction at $t = 0$ is completely free of thorium; i.e. $N_2^0 = 0$. However, after some time it is possible to detect the presence of ^{234}Th. The change in the number of ^{234}Th with time follows the first term of (4.54); in fact, in Figure 1.1 the measurements detect only the ^{234}Th nuclide (β-emitting), since the detection system used is not sensitive to the α's from ^{238}U ($\psi_\alpha = 0$). The time of observation is much smaller than the half-life of the ^{238}U decay, so there will be no observable change in the number of atoms of uranium during the time of observation, i.e. $N_1 = N_1^0$. Further, since $t_{½}$ for $^{238}U \gg t_{½}$ for ^{234}Th, i.e. $\lambda_1 \ll \lambda_2$, we can simplify (4.54) to

$$N_2 = (\lambda_1 /\lambda_2) N_1 (1 - e^{-\lambda_2 t}) \tag{4.55}$$

According to this equation, the number of ^{234}Th atoms, N_2, increases with time with the half-life of ^{234}Th. In other words, after a period of 24.1 d there is 50% of the maximum value of ^{234}Th, after 48.2 d there is 75% of the final maximum value, etc. This is illustrated by the change in the uranium fraction activity in Figure 1.1. Further, from the relationship (4.55) we can see that the maximum value of thorium ($t = \infty$) is given by

$$N_2 \lambda_2 = N_1 \lambda_1 \tag{4.56}$$

These equations, based on $\lambda_1 \ll \lambda_2$, show that the amount of daughter atoms becomes constant after some time. At that time the rate of decay of the daughter becomes equal to

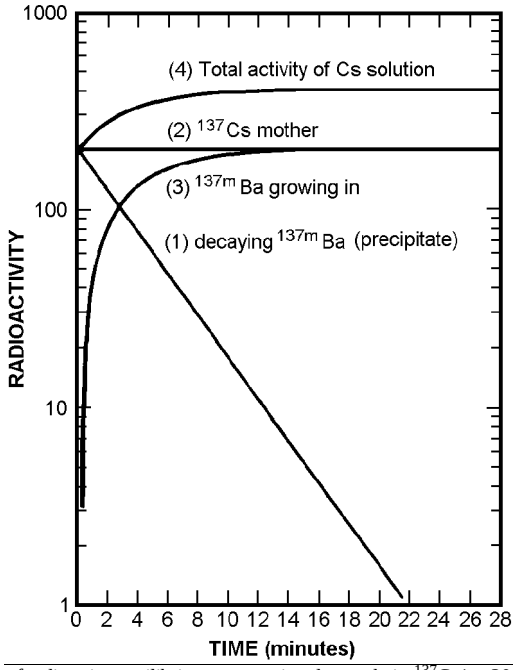


FIG. 4.11. Case of radioactive equilibrium: successive decay chain $^{137}Cs(t_{½}$ 30 y) → $^{137m}Ba(t_{½}$ 2.6 min) → stable.

the rate of decay of the parent, i.e. $A_2 = A_1$, but the amounts of the parent and the daughter are not equal since N_2 is much smaller than N_1. This condition of $A_2 = A_1$ is known as *secular equilibrium*, which is a misnomer since this is a steady state and not a true equilibrium situation. It is also common to speak of *radioactive equilibrium* in referring to this steady state condition.

We can calculate that at secular equilibrium for each gram of ^{238}U there will be present 1.44×10^{-11} g of ^{234}Th and 4.9×10^{-16} g of ^{234}Pa. Since the specific radioactivity of ^{238}U is 746 000 dpm/g, the decay rate of 4.9×10^{-16} g ^{234}Pa is also 746 000 dpm.

When the time of observation is very short compared to the half-life of the parent nuclide, as in secular equilibrium, no change in the decay rate of the parent is observed for many daughter half-lives. Our example of ^{137}Cs, which decays via the isomeric state ^{137m}Ba to ^{137}Ba, presents another case of "secular equilibrium". If we have an "old" sample in which radioactive equilibrium has been reached (older than ~15 min, since the $t_{½}$ of the daughter is 2.6 min), and separate the cesium from the barium by precipitation and filtration of $BaSO_4$, the activity measured from the precipitate will follow curve (1) in Figure 4.11. In the filtrate solution the activity from ^{137}Cs, curve (2), is unchanged during our observation time. However, ^{137m}Ba grows into the solution, curve (3), so that the total activity of the solution, curves (2) plus (3), increases according to curve (4).

In many radioactive decay chains the half-life of the parent is longer than that of the daughter but it is short enough that a change in the disintegration rate of the parent is observable during the period of observation of the experiment. In such cases the system

reaches the condition termed *transient equilibrium*. The length of time of observation of the activity of the sample may be the determining factor as to whether it appears to be transient or secular equilibrium. If a parent has a one month half-life and the observation of the change in decay rates of parent and daughter extends over an hour or even a few days, the data would follow the equation for secular equilibrium since the degree of change in the parent decay would be negligible. However, if the observation extends over a period of several weeks or months, then the change in the decay rate of the parent is significant and it would appear as transient equilibrium.

The case of transient equilibrium will be illustrated by an example, such as the decay chain

$$^{140}\text{Ba}(\beta^-,\ 12.75\ \text{d})\ ^{140}\text{La}(\beta^-,\ 1.678\ \text{d})\ ^{140}\text{Ce}(\text{stable})$$

^{140}Ba is one of the most important fission products. If we isolate barium, lanthanum grows into the sample. Figure 4.12 shows the decay of ^{140}Ba in curve (1), which follows the simple decay of (4.41). Curve (2) shows the activity of the daughter, for which the left half of eq. (4.54) is valid. Replacing decay constants by half-lives we can rewrite this equation as

$$A_2 = t_{½,1}/(t_{½,1} - t_{½,2})A_1^0(e^{-0.693t/t_{½,1}} - e^{-0.693t/t_{½,2}}) \tag{4.57}$$

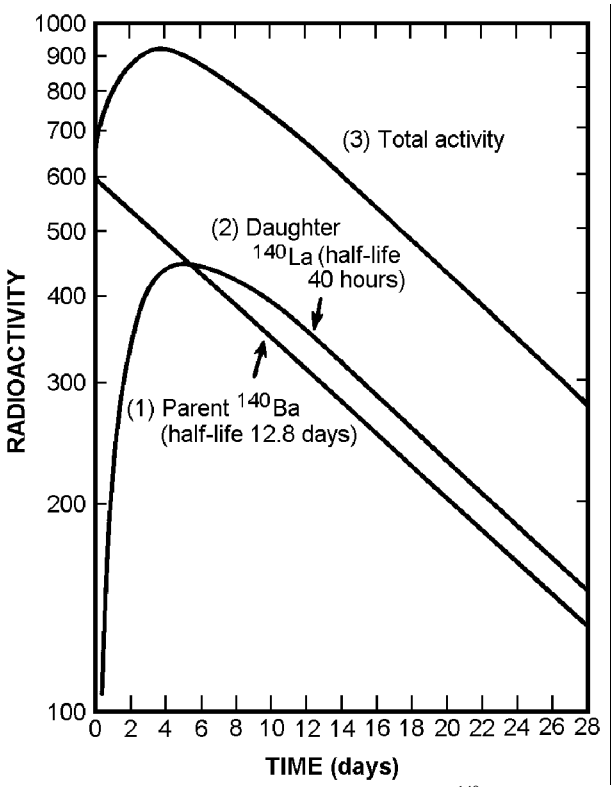


FIG. 4.12. Case of transient equilibrium: successive decay chain ^{140}Ba($t_{½}$ 12.75 d) → ^{140}La($t_{½}$ 1.678 d) → stable.

At $t \ll t_{½,1}$ ($t \ll 12.8$ d) the first exponential term is very close to 1 and A_2 increases proportional to $(1 - e^{-0.693t/t_{½,2}})$; this is the increasing part of curve (2). At $t \gg t_{½,2}$ ($t \gg 40$ h), the second exponential term becomes much smaller than the first one, and A_2 decreases proportional to $e^{-0.693t/t_{½,1}}$. For this part of the curve we may write

$$N_2 = N_1 \lambda_1 / (\lambda_2 - \lambda_1) \tag{4.58}$$

which is the relation valid for transient equilibrium. The total activity of the barium sample, curve (3), is the sum of curves (1) and (2).

If the parent is shorter-lived than the daughter, the daughter activity grows to some maximum value and then decays with its own characteristic half-life. This contrasts to the case of transient equilibrium where the daughter has an apparent decay given by the half-life of the parent. An example of this is shown in Figure 4.13 for the decay chain

$$^{218}\text{Po}(\alpha,\ 3\ \text{min})\ ^{214}\text{Pb}(\beta^-,\ 27\ \text{min})\ ^{214}\text{Bi}$$

The time necessary for obtaining the maximum daughter intensity in the non-equilibrium case of the shorter-lived parent is given by

$$t_{max} = (\lambda_2 - \lambda_1)^{-1} \ln(\lambda_2/\lambda_1) \tag{4.59}$$

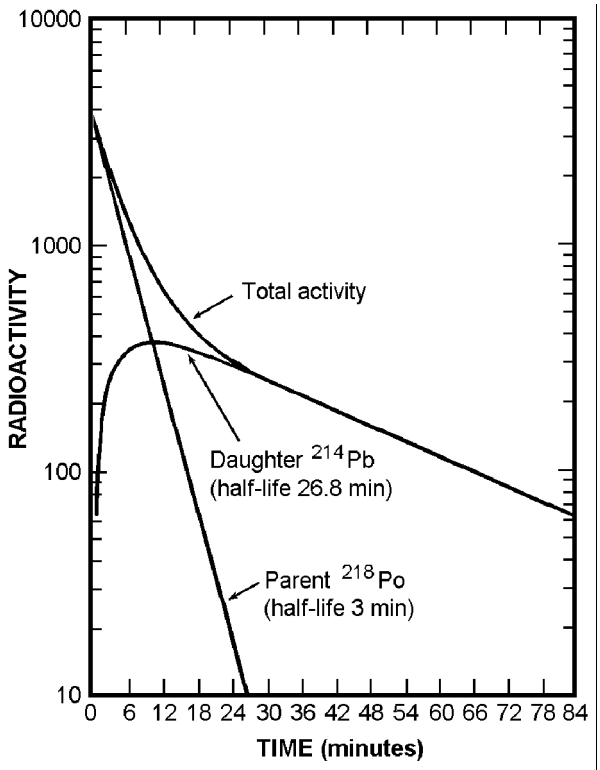


FIG. 4.13. Case of no equilibrium: successive decay chain $^{218}\text{Po}(t_{½}\ 3\ \text{min}) \rightarrow {}^{214}\text{Pb}(t_{½}\ 26.8\ \text{min}) \rightarrow$ stable.

4.16. Radioisotope generators

The growth of radioactive daughters frequently has practical significance. For example, in radiation therapy and diagnostic medicine it is preferable to use short-lived nuclides. In fact, it is preferable to conduct tracer experiments with short-lived nuclides as this eliminates the problem of disposal of residual radioactive waste after completion of the experiment. It is convenient to have a long-lived mother in storage from which a short-lived daughter can be removed as required for use in tracer work. A few examples of uses of such mother-daughter pairs are discussed; others are included in Table 4.1.

Such systems are called *radioisotope generators*. ^{222}Rn is sometimes used for the radiotherapeutic treatment of cancer. This product is isolated by separating it as a gas from the parent substance ^{226}Ra which is normally in the form of solid or a solution of $RaBr_2$. ^{222}Rn grows into the radium sample with a half-life of 3.8 d. After a 2-week period, following a separation of radon from radium, approximately 90% of the maximum amount of radon has grown back in the radium sample. Consequently, it is useful to separate ^{222}Rn each 2 weeks from the radium samples since further time provides very little additional radioactivity. The ^{222}Rn is an α emitter; the therapeutic value comes from the irradiation of the tissue by the γ-rays of the decay daughters ^{214}Pb and ^{214}Bi which reach radioactive equilibrium extremely rapidly with the ^{222}Rn.

^{99m}Tc is used for diagnostic purposes for liver, spleen, and thyroid scanning (§9.5). The ^{99}Mo parent, obtained by chemical separation from ^{235}U fission product mixture, is absorbed on a column of alumina and the daughter ^{99m}Tc removed by passage of saline solution at intervals governed by the equilibrium. The parent, when it is fixed in a semipermanent sample as on an adsorbent column, is often known as a cow and the removal of the daughter activity from the

Table 4.1. *Some common radioactive milking pairs. The decay properties include decay energy (MeV), mode of decay and half-life*

Mother nuclide	Decay properties	Daughter nuclide	Decay properties	Application
^{44}Ti	EC, γ; 47.3 y	^{44}Sc	1.5 β^+; 1.16 γ; 3.93 h	Teaching
^{68}Ge	EC; 270.8 d	^{68}Ga	1.9 β^+; 1.08 γ; 1.135 h	Medical
^{87}Y	EC; 3.35 d	^{87m}Sr	0.39 γ; 2.80 h	Medical & teaching
^{90}Sr	0.5 β^-; 28.5 y	^{90}Y	2.3 β^-; 2.671 d	Heat source†, Calibration source
^{99}Mo	β^-, γ; 65.9 h	^{99m}Tc	0.14 γ; 6.0 h	Medical
^{113}Sn	EC, γ; 115.1 d	^{113m}In	0.39 γ; 1.658 h	Medical
^{132}Te	β^-, γ; 78.2 h	^{132}I	2.1 β^-, γ; 2.28 h	Medical
^{137}Cs	β^-, γ; 30.0 y	^{137m}Ba	0.66 γ; 2.55 m	Gamma radiography, Radiation sterilization†
^{140}Ba	β^-, γ; 12.75 d	^{140}La	β^-, γ; 1.678 d	Lanthanum tracer
^{144}Ce	β^-, γ; 284.9 d	^{144}Pr	3.0 β^-; 17.28 m	Calibration source
^{210}Pb	β^-, γ; 22.3 y	^{210}Bi	1.2 β^-; 5.01 d	Calibration source
^{226}Ra	α; 1600 y	^{222}Rn	α; 3.825 d	Medical
^{238}U	α; 4.468 x 10^9 y	^{234}Th	β^-, γ; 24.1 d	Thorium tracer

† Main use of mother substance in large amounts….

radioisotope generator (the "cow") is termed *milking*.

Another commonly used radioisotope generator is ^{132}Te from which ^{132}I may be milked. In this case ^{132}Te is adsorbed as barium tellurite on an alumina column, and the ^{132}I removed by passage of 0.01 M ammonia through the column. The ^{132}I is used both diagnostically and therapeutically for thyroid cancer.

Many of these sources produce radionuclides with half-lives suitable for teaching purposes, e.g. ^{137m}Ba(2.6 min), ^{144}Pr(17.3 min), ^{44}Sc(4.0 h), ^{99m}Tc(6.0 h) and ^{90}Y(64 h).

4.17. Decay energy and half-life

It was observed early in both α- and β-decay that the longer the half-life the lower the decay energy. Although there are many exceptions to this observation H. Geiger and J. M. Nuttall formulated the law

$$\log \lambda_\alpha = a + b \log R_{\text{air}} \tag{4.60}$$

for the natural α-active nuclides. Here a and b are constants, and R_{air} is the range of the α-particles in air which is directly proportional to the α-particle energy E_α. A similar relation was deduced by E. Fermi for the β-decay:

$$\log \lambda_\beta = a' + b' \log E \tag{4.61}$$

where a' is a constant related to the type of β decay and $b' \approx 5$.

Although these rules have been superseded by modern theory and the enormous amount of nuclear data now available, they may nevertheless be useful as rough guides in estimates of half-lives and decay energies. In §11.7 more valid but more complicated relationships are discussed.

4.18. The Heisenberg uncertainty principle

In this chapter we have repeatedly stated that the nuclear decay energies are exact values as required by quantum mechanics. However, this is not exactly correct: the energy levels have a certain "spread". This was first stated by Heisenberg in 1927 and is of fundamental importance in all areas of nuclear physics.

The *uncertainty principle* states that it is impossible to measure simultaneously the exact position and the exact momentum of a particle. This follows from the wave properties of the particle. If for example, we attempt to measure the exact position of an electron by observing the light emitted when it hits a scintillating screen, this act interferes with the movement of the electron causing it to scatter, which introduces some uncertainty in its momentum. The size of this uncertainty can be calculated exactly and is related to the Planck constant. If Δx denotes the uncertainty in position and Δp the uncertainty in momentum along the x axis, then

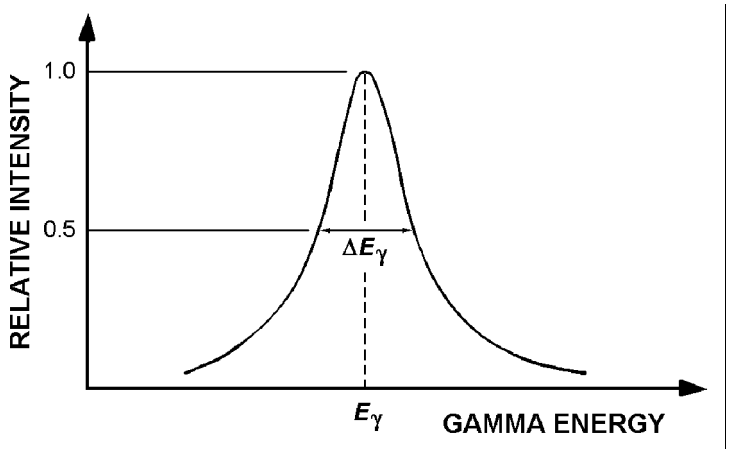


FIG. 4.14. The energy half-width value ΔE_γ is the FWHM of the γ-peak.

$$\Delta x \, \Delta p \geq \mathbf{h}/(2\pi) \equiv \tag{4.64}$$

is called "h-bar" (1.05 x 10^{-34} J s), and **h** is the *Planck constant*.

This principle holds for other *conjugate variables*, as angle θ and angular momentum p_θ

$$\Delta\theta \, \Delta p_\theta \geq \tag{4.65}$$

and time and energy

$$\tau \, \Delta E \geq \tag{4.66}$$

This latter equation relates the life-time τ of an elementary (or nuclear) particle to the uncertainty in its energy (ΔE). For excited nuclear states this can be taken as the width of the γ-peak at half-maximum intensity (the "FWHM value") (Fig. 4.14). For example, if ΔE_γ = 1.6 keV, then $\tau \geq 1.05 \times 10^{-34}/1600 \times 1.60 \times 10^{-19}$ s = 4.1×10^{-19} s. This is a long time compared to that of a nuclear rotation, which is about 10^{-23} s. Consequently the wave mechanic properties of particles (and γ-rays) introduces a certain fundamental uncertainty in the particle energy. Several applications of this are described in later chapters.

4.19. Exercises

For some of the problems necessary nuclear data are given in the Tables or appendices.

4.1. ^{239}Pu emits α-particles of maximum 5.152 MeV. What is the recoil energy of the product formed?

4.2. Using a magnetic spectrometer the maximum energy of the electrons from ^{137}Cs was found in Figure 4.1 to correspond to 3.15×10^{-3} Tesla m. Calculate the energy (a) assuming that the electrons are non-relativistic, (b) with correction for relativistic mass increase.

4.3. ^{11}C decays through emission of positrons of a maximum energy of 1.0 MeV. Calculate the recoil energy of the daughter.

4.4. ^{16}N decays through β^- decay to ^{16}O with a half-life of 7.1 s. A number of very energetic γ's follow after the β-emission, the dominating one with an energy of 6.14 MeV. What is the ^{16}O recoil energy?

4.5. The binding energy of a K-electron in barium is 37 441 eV. Calculate from Figure 4.1 the internal conversion energy for ^{137m}Ba (Fig. 4.5).

4.6. From the specific activity of potassium (1850 dpm/g K) and the fact that it all originates in the rare isotope ^{40}K (0.0117%), calculate the half-life of ^{40}K.

4.7. One may assume that when ^{238}U was formed at the genesis an equal amount of ^{235}U was formed. Today the amount of ^{238}U is 138 times the amount of ^{235}U. How long a time ago did the genesis occur according to this assumption?

4.8. The interior of the earth is assumed to be built up of a solid core (radius 1371 km) followed by a molten core (radius 3471 km) and a molten mantle (radius 6354 km) covered by a 17 km thick crust. One assumes that 2% by weight of the molten mantle and crust is potassium; the average mantle density is assumed to be 6000 kg m^{-3} and that of the crust 3300 kg m^{-3}. What energy outflow will the radioactive decay of this element cause at the earth's surface? The decay scheme of ^{40}K is given in eqn. (5.5). For the EC branch $Q = 1.505$ MeV, for the β^- branch 1.312 MeV. Each decay by the EC branch leads to emission of a 1.46 MeV γ. Compare this energy output to the solar energy influx to the earth of 3.2×10^{24} J y^{-1}.

4.9. A hospital has a 1.5 Ci source of ^{226}Ra in the form of a $RaBr_2$ solution. If the ^{222}Rn is pumped out each 48 h, what is (a) the radon activity (in Bq) at that moment, (b) the radon gas volume at STP?

4.10. (a) Prove the correctness of eqn. (4.20) by using Newton's laws of motion. (b) Prove the correctness of eqn. (4.34).

4.11. A recently prepared ^{212}Pb sample has the activity of 10^6 dpm. (a) What is the activity 2 h later? (b) How many lead atoms are left in the sample at this moment? $t_{½}$ 10.64 h.

4.12. A radioactive sample was measured at different time intervals:

Time (h)	Activity (dpm)	Time (h)	Activity (dpm)
0.3	11100	30	1015
5	5870	40	888
10	3240	50	826
15	2005	100	625
20	1440		

Determine the half-lives of the two nuclides (not genetically related) in the sample and their activities (in Bq) at time $t = 0$. The background of the detection device was 100 376 counts per 1 000 min; its counting efficiency was 17%.

4.13. The α-activity of a mixture of astatine isotopes was measured at different times after their separation, giving the following results:

t(min)	A(dpm)	t(min)	A(dpm)
12	756	121	256
17.2	725	140	215.5
23.1	638	161	178.5
30.0	600	184	150.7
37.7	545	211	127.3
47.5	494	243	101.9
59.5	435	276	84.9
73	380	308	68.2
87	341	340	55.0
102	288		

Calculate the half-lives of the isotopes and their activities at $t = 0$.

4.14. In the ion source of a mass spectrograph, UF_6 vapor is introduced which partly becomes ionized to UF_5^+. The ionic currents were measured at mass positions 333, 330, and 329.The ion current ratios were $I_{333}/I_{330} = 139$, and $I_{330}/I_{329} = 141.5$. What is the half-life of ^{234}U if that of ^{238}U is 4.5×10^9 y? Radioactive equilibrium is assumed to exist in the UF_6.

4.20. Literature

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