CHAPTER 2

NUCLEAR PROCESSES

The control of nuclear phenomena represents the fundamental aspect of nuclear energy conversion. It is our intent here to provide a summary description of some of those nuclear processes which are of particular relevance in the analysis of nuclear reactor systems.

2.1 SOME PROPERTIES OF NUCLEI

An atom may be visualized as an entity consisting of a positively charged nucleus surrounded by a negatively charged electron cloud. Although the electrons are important in chemical reactions and in charged particle transport, they are of secondary importance in nuclear reactors.

As indicated previously, a nucleus is identified by its constituent nucleons which consist of positively charged protons and uncharged neutrons. The following symbols and definitions are widely used:

Z = number of protons (atomic number or proton number),
N = number of neutrons (neutron number),
A = Z + N (mass number).

A specific nucleus is identified by ^{A}X where X is the abbreviation of the name of the element. Figure 2.1 provides a graphical representation of some selected nuclei.

The mass of a proton, m_p, is almost identical to the mass of a neutron, m_n; these are given in terms of grams (g) and atomic mass units (amu) by

$$m_p = 1.675 \times 10^{-24} \text{ g} = 1,0073 \text{ amu},$$

$$m_n = 1.602 \times 10^{-24} \text{ g} = 1.0087 \text{ amu}.$$

The radius of a nucleus may be approximated by the formula

$$R = 1.25 \times 10^{-13}(A)^{1/3} \text{ cm}, \qquad (2.1)$$

where A is the atomic mass number of the nucleus of interest. Using aluminum as a convenient example we find that its nuclear radius is equal to

$$R_{Al} = 1.25 \times 10^{-13} \times (27)^{1/3} = 3.75 \times 10^{-13} \text{ cm.} \qquad (2.2)$$

and therefore can be viewed as occupying a volume of

$$V_{Al} = \frac{4}{3}\pi(R_{Al})^3 = 1.24 \times 10^{-37} \text{ cm}^3 . \qquad (2.3)$$

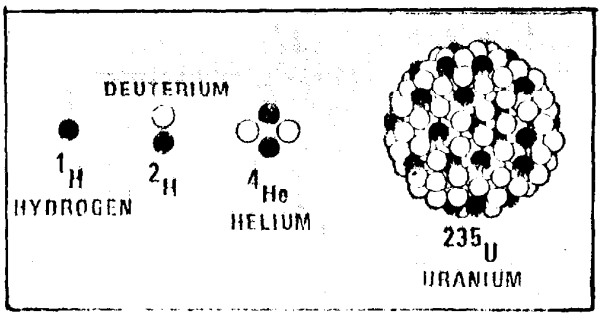

FIG. 2.1: Simplified visualization of the hydrogen nucleus, the deuterium nucleus, a helium nucleus,and an uranium nucleus. The black spheres represent protons while the white spheres represent neutrons.

The number of atoms per unit volume, N, may be determined with the aid of the material density, ρ, Avogadro's relationship involving the number of atoms in a mole, and the atomic number of the substance. This atomic density is given by

$$N = \frac{\rho(\text{g/cm}^3)N_A(\text{atoms/mole})}{A(\text{g/mole})} , \qquad (2.4)$$

where A is equal to the atomic mass number and N_A is Avogadro's number given by

$$N_A = 0.602 \times 10^{24} \text{ atoms/mole} .$$

Again, for aluminum we obtain

$$N = \frac{2.7 \times 0.602 \times 10^{24}}{27} = 6.02 \times 10^{22} \text{ atoms/cm}^3 . \qquad (2.5)$$

The above sample calculations indicate the relative change in scale from the more traditional numerical description of macroscopic phenomena.

2.2 MASS DEFECT AND BINDING ENERGY

Recalling our mechanistic picture of the nucleus as consisting of individual nucleons, it is known that the total mass of separate protons and neutrons exceeds the mass of the nucleus consisting of these nucleons. This mass difference Δm is given in terms of the total mass of individual protons, Zm_p, the total mass of individual neutrons, Nm_n, and the mass of the nucleus made up of these $A = Z + N$ protons and neutrons, m_A:

$$\Delta m = Zm_p + Nm_n - m_A \ . \tag{2.6}$$

When this mass defect is substituted into Einstein's mass-energy relation

$$E = (\Delta m)c^2 \ , \tag{2.7}$$

we obtain the energy with which the nucleus is held together. This is called, appropriately, the binding energy of the nucleus ${}^{A}X$ and represented by the symbol B.E.

The evaluation of the binding energy, Eq. (2.7), can be undertaken with any set of consistent units for mass and for the speed of light. For example, in SI units and for a one kilogram mass we obtain

$$E = 1(\text{kg}) \times 2.998 \times 10^8 (\text{m/s})^2 = 8.998 \times 10^{16} \text{ J} = 2.5 \times 10^7 \text{ MWh}. \tag{2.8}$$

In practice it has been found convenient to use a unit of energy defined as an electron volt (eV) and its multiples, kiloelectron volts (keV), and million electron volts (MeV). In the calculation of mass defects it can be shown that, with the use of appropriate conversion constants, one atomic mass unit of mass is equivalent to 931 MeV of energy:

$$E = (1 \text{ amu})c^2 = 931 \text{ MeV} \ . \tag{2.9}$$

With this equivalence and our previous expressions we may calculate the binding energy of, say Uranium-235:

$$\Delta m_u = Zm_p - Nm_n - m_A \ ,$$

$$= (92 \times 1.0073) + (143 \times 1.0087) - (235.0439) = 1.872 \text{ amu}. \tag{2.10}$$

This is equivalent to 1742.832 MeV. The binding energy per nucleon B.E./A, for Uranium-235 is hence given by

$$\text{B.E./A} = \frac{1742.832}{235} = 7.416 \text{ MeV/A} \ . \tag{2.11}$$

Figure 2.2 shows the binding energy per nucleon for all known stable nuclei.

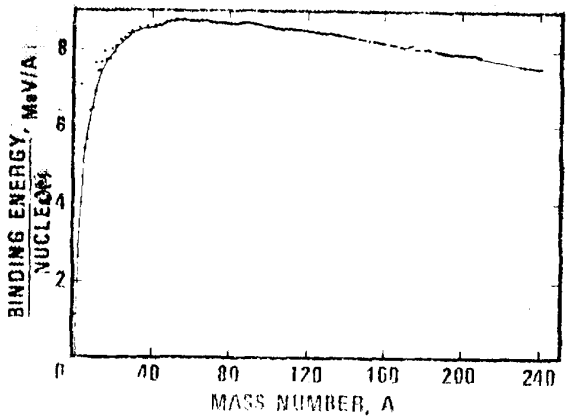

FIG. 2.2: Binding energy per nucleon for the known stable nuclei.

The energy which becomes available in a fission process represents the difference in the binding energy of the nucleus which undergoes fission and the total binding energies of all subsequent fission product nuclei. In principle, this can be calculated for any one of the numerous reaction products possible. By reference to Figure 2.2 we can undertake a quick calculation to obtain a typical order of magnitude. The value of binding energy per nucleon for uranium is about 7.4 MeV/A. Since most fission products attain an atomic mass of approximately 95 and 140 we estimate that the effective binding energy per nucleon, Fig. 2.2, after fission must be about 8.3 MeV/A. This represents a net gain in binding energy of 8.3 - 7.4 = 0.8 MeV/A. The total energy release therefore is of the order of 235 x 0.9 MeV/A = 212 MeV per fission. As indicated earlier an excess of 80% of this energy appears in the form kinetic energy of the fission products and thus represents potentially recoverable energy.

2.3 RADIOACTIVITY AND FISSION PRODUCTS

All nuclei possess an internal energy level structure. Under radiation conditions which occur continually in a nuclear reactor, there exist numerous processes whereby a nucleus can become excited and attain one of these higher energy levels. Indeed, most of the fission products exist initially in an excited state. Figure 2.3 illustrates the nuclear level structure for light, intermediate, and heavy nuclei.

The attainment of an excited nuclear level is commonly a temporary condition. Generally there exist several possibilities whereby the excited nucleus can eventually attain the stable, ground state. It may, for example, eject a nucleon, or the excited nucleus may eject a beta particle, or it may capture an electron; further, it may also emit a gamma ray and thus attain a lower energy

level. We illustrate the possibilities for radioactive Iron-59, Figure 2.4.

Whatever the decay scheme may be, the important feature to note is that radiation is emitted when a nucleus decays. This represents a potential health hazard and requires that both the rate of radioactive decay and the type and energies of radiation emitted be considered. We will consider the rate of radioactive decay here; the type and energy of radiation emitted for some nuclei will be referred to but a more comprehensive listing can be found by reference to any one of many tabulations.

The rate of decay of an ensemble of identically excited nuclei follows a well defined time dependence and illustrates a characteristic property of nuclear states. Supposing that at time t there exist N(t) excited nuclei in a unit volume. The fractional decrease in the number of the excited nuclei is directly proportional to the time interval of observation. That is

$$-\frac{\Delta N}{N(t)} \propto \Delta t. \qquad (2.12)$$

Introducing a proportionality constant λ, yields

$$-\frac{\Delta N}{N(t)} = \lambda \Delta t. \qquad (2.13)$$

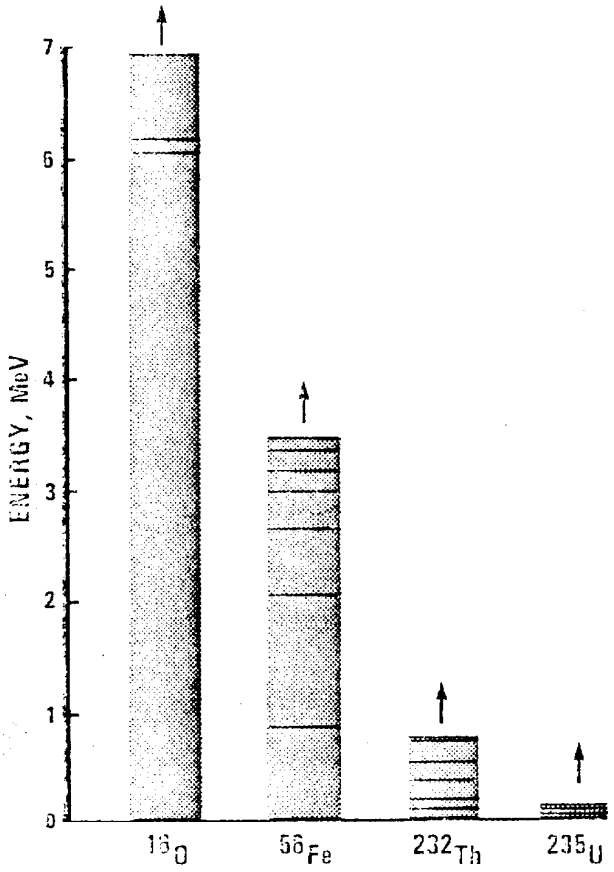

FIG. 2.3: Graphical visualization of the nuclear energy level structure of several nuclei of various masses. Note that the levels of the heavier nuclei are more close spaced and lower than for the lighter nuclei. The ground state of the nucleus corresponds to zero energy.

Taking the limit of $\Delta N/\Delta t \to dN/dt$ leads to a differential equation which may be integrated directly. The result can be shown to be given as

$$N(t) = N(0)\exp[-\lambda t] \; . \qquad (2.14)$$

That is, the number of radioactive nuclei decreases exponentially with time. The decay constant λ thus represents the parameter which specifies how quickly the concentration of a particular radioactive nuclei decreases with time; it has been measured and tabulated for all radioactive nuclei of interest in nuclear reactors.

For many practical purposes it has been found more useful to refer to the half-life, $T_{1/2}$, of a particular radioactive nuclide rather than to its decay constant; this term refers to the time required for the concentration of a radioactive specie to be reduced by a factor of one-half; it can be shown that this parameter is a constant and is determined as follows. Using t = 0 as the initial time of interest, we wish to find $T_{1/2}$ for which

$$N(T_{1/2}) = \frac{1}{2} N(0). \qquad (2.15)$$

Using Eq. (2.14) permits us to equate

$$\frac{1}{2} N(0) = N(0)\exp[-\lambda T_{1/2}]. \qquad (2.16)$$

Cancelling N(0) and solving for $T_{1/2}$ yields

$$T_{1/2} = \frac{\ell n(2)}{\lambda} \simeq \frac{0.6931}{\lambda} . \qquad (2.17)$$

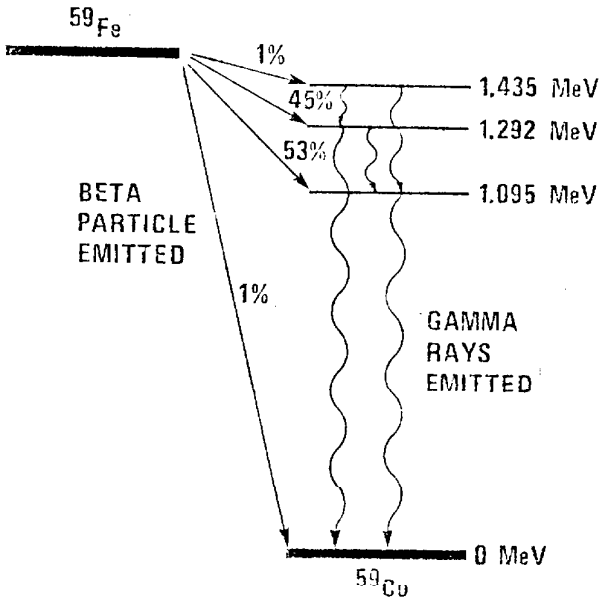

FIG. 2.4: Radioactive decay of Iron-59 to Cobalt-60. The straight-line arrows denote beta decay and the wavy arrows denote gamma ray emission as the Cobalt nucleus attains a lower energy level.

Figure 2.5 provides a graphical representation of the decay of an ensemble of radioactive nuclei where time is expressed in units of half-life.

In many circumstances, it is equally important to know the rate at which radioactive nuclei decay. This disintegration rate is given by differentiation of Eq. (2.14) with respect to time:

$$-\frac{dN(t)}{dt} = \lambda N(0)\exp[-\lambda t] = \lambda N(t). \qquad (2.18)$$

Or, using Eq. (2.17)

$$-\frac{dN(t)}{dt} = \frac{0.6931\ N(t)}{T_{1/2}} . \qquad (2.19)$$

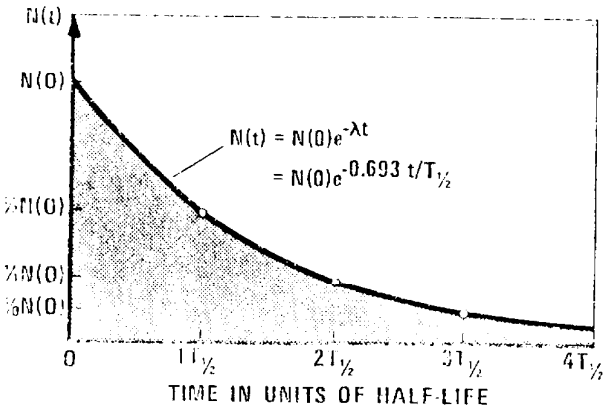

FIG. 2.5: Exponential decrease with time of the number of radioactive nuclei. N(0) identifies the number of radioactive nuclei existing initially.

The negative sign appears because we are describing a particle density which decreases with time. The name Curie, abbreviated Ci, has been officially adopted as a unit of disintegration rate and is defined by

$$1 \text{ Ci} = 2.7 \times 10^{10} \text{ dps} \qquad (2.20)$$

where dps represents disintegrations per second. This parameter, together with the type and energy of the emitted radiation is of fundamental importance in the determination of radiation exposure standards.

In the study of radioactivity and radioactive nuclei, it is necessary to distinguish between natural radioactivity and artificial or man-made radioactivity. The naturally occurring radionuclides can be grouped in the following categories:

Group #1: Primary radioactive nuclides possessing half-lives on the geological time scale. Some of these nuclides and their half-lives are listed in Table 2.1

Group #2: Decay products of the above which are again radioactive.

Group #3: Induced radionuclides produced by cosmic radiation. The nucleus Carbon-14 is one such example.

We point out that many of the radionuclides in Groups #1 and #2 appear in the earth's crust and appear commonly in building products such as concrete and stone; Potassium-40, of course, exists in the human body.

The number of radioactive nuclides occurring in a nuclear reactor as a result of the fission process is very large indeed. These radionuclides can be grouped from the standpoint of radioactive waste management. The groupings are as follows:

Group #1: Those which after one year of decay may, if desired, be released because they have only stable nuclides or they are of such long half-lives that they occur in nature.

Group #2: Those which may be released after a further 9 years of decay.

Group #3: Those which could be released after 2,000 years storage fused in glass blocks or the equivalent.

Group #4: Those requiring indefinite retention or special management.

Nuclide	Half-Life
^{40}K(Potassium-40)	1.3×10^{9} yrs.
^{87}Rb(Rubidium-87)	5.0×10^{10} yrs.
^{115}In(Indium-115)	6.0×10^{14} yrs.
^{147}Sm(Samarium-147)	1.0×10^{11} yrs.
^{232}Th(Thorium-232)	1.4×10^{10} yrs.
^{235}U(Uranium-235)	7.1×10^{8} yrs.
^{238}U(Uranium-238)	4.5×10^{9} yrs.

TABLE 2.1: Tabulation of some long lived radionuclides which exist naturally.

We now consider a more systematic discussion of nuclear transformations.

2.4 NUCLEAR TRANSFORMATION

One nuclear decay process which involves the emission of a gamma ray only represents a case whereby an excited nucleus, say $(^{A}X)^{*}$, attains its ground state, ^{A}X. Symbolically, we write

$$(^{A}X)^{*} \rightarrow {}^{A}X + \gamma + Q, \tag{2.21}$$

here γ represents the gamma ray photon and Q represents the energy released in the process; this Q-value includes the recoil energy of the nucleus ^{A}X and the energy of the γ-ray.

The analysis of other radioactive decay processes and more complex nuclear transformations is enhanced by the use of a graphical listing of all known stable and unstable nuclei. One such representation is the Chart of the Nuclides which lists each nucleus and some of its properties on a N-Z Cartesian coordinate system. Figure 2.6 shows a portion of such a table for the light elements.

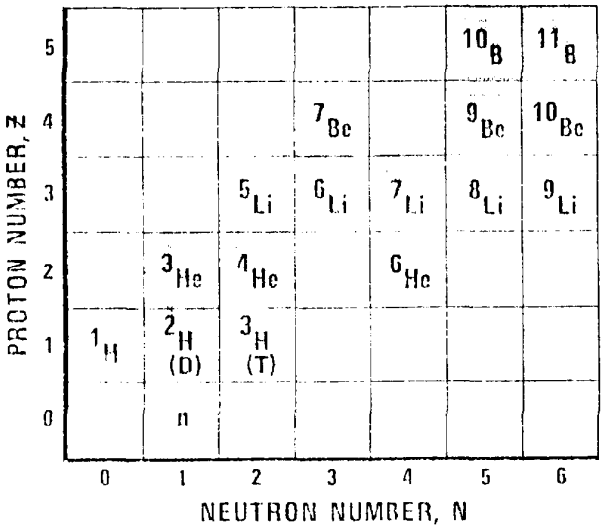

FIG. 2.6: Portion of the Chart of the Nuclides showing the lighter elements. The nuclides denoted by shaded squares are stable whereas the others are known unstable (radioactive) nuclides.

As an example of a specific nuclear transformation we consider the naturally occurring radioactive specie Carbon-14 which is known to decay to Nitrogen-14 by the emission of a beta particle β^-. This process, called beta decay, may be written as

$$^{14}C \rightarrow {}^{14}N + \beta^- \ . \qquad (2.22)$$

No gamma radiation is involved because both Carbon-14 and Nitrogen-14 appear only in their ground state; for reasons of clarity we have not shown the Q-value in the particle-balance equation. In general, such a decay process is represented by

$$^{A}X \rightarrow {}^{A}Y + \beta^- \ , \qquad (2.23)$$

where the transformation from element X with Z protons and N neutrons proceeds to an element Y with (Z+1) protons and, to preserve A, (N-1) neutrons. Beta decay processes are particularly important in the breeding of fissile material.

Associated with every beta decay is the concurrent transformation

$$n \rightarrow p + \beta^- \quad , \tag{2.24}$$

where n is a neutron and p a proton. Although this transformation occurs in beta decay as part of the nuclear process, it is known that free neutrons are unstable and decay according to Eq. (2.24) with a half-life of 11.3 minutes.

An alpha decay process is similarly represented by

$$^{A}X \rightarrow {}^{A-4}Y + He^4 \quad . \tag{2.25}$$

The product ^{A-4}Y clearly possesses two fewer protons than ^{A}X because nucleons must be conserved.

The above as well as other decay processes may be conveniently represented on the Chart of Nuclides in a graphical form. This is shown in part of Fig. 2.7.

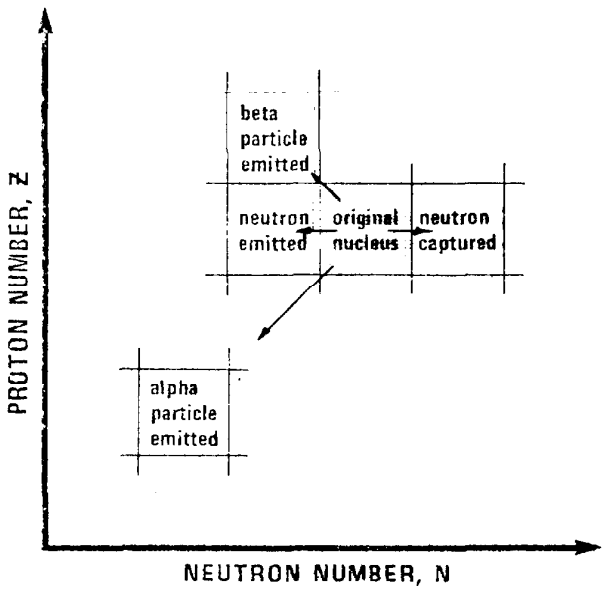

FIG. 2.7: Graphical representation of nuclear transmutation associated with beta emission, neutron emission, alpha emission and neutron capture. The original nucleus is denoted by the shaded area.

In addition to the decay processes which lead to transformations, there exist important induced nuclear transformations. Some of these may be produced by accelerating ions and nuclear particles using nuclear accelerators while others,

such as the neutron absorption process, occur in a nuclear reactor environment. Of particular interest in the analysis of nuclear reactors is the neutron induced transformation and the decay of the reaction products. For example, a neutron capture process is written

$$ {}^{A}X + n \rightarrow {}^{A+1}X \quad . \tag{2.26} $$

The nucleus ${}^{A+1}X$ may or may not decay by any one of the various decay processes; the decays observed are governed entirely by the statistical properties of competing nuclear-decay possibilities. For the fission process we may write explicitly

$$ {}^{A+1}X \rightarrow (FP)_1 + (FP)_2 + \nu n \quad , \tag{2.27} $$

where $(FP)_1$ and $(FP)_2$ identify two fission products and ν specifies the number of neutrons emitted. Other forms of radiation which might appear concurrently are not shown. Thus, for the neutron induced fission process in Uranium-235 we may write as one possible reaction

$$ {}^{235}U + n \rightarrow {}^{236}U \rightarrow {}^{95}Mo + {}^{139}La + 2n \quad . \tag{2.28} $$

The Q-value can be calculated to be 208 MeV. The total number of neutrons and protons is conserved in each of these three stages. The time required for this entire process is of the order of 10^{-14} sec.

Another important neutron induced transformation, whereby a neutron is permanently removed, is the capture process in xenon:

$$ {}^{135}Xe + n \rightarrow {}^{136}Xe \quad . \tag{2.29} $$

Although Xenon-135 is a radioactive specie which appears as a fission product, it may decay before neutron capture can occur; the product nucleus Xenon-136, however, is stable.

As a final example of a neutron induced process, we indicate the process whereby neutron capture in a stable non-fissile nucleus can potentially lead to a fissile nucleus by a series of radioactive decay processes. We consider the element thorium which appears naturally as the isotope Thorium-232 with 100% abundance. The neutron capture in Thorium-232 is written as

$$ {}^{232}Th + n \rightarrow {}^{233}Th \quad . \tag{2.30} $$

The product nucleus is radioactive and decays to Protactinium-233 by beta decay

$$ {}^{233}Th \rightarrow {}^{233}Pa + \beta^- \quad , \tag{2.31} $$

with a half-life of 22.1 minutes. The product Protactinium-233 is also unstable and decays to fissile Uranium-233 with a half-life of 27.4 days:

$$ {}^{233}Pa \rightarrow {}^{233}U + \beta^- \quad . \tag{2.32} $$

Thus, the non-fissile nucleus Thorium-232 has been transmuted into the important fissile nucleus Uranium-233 which can contribute to nuclear energy production. This entire sequence may be conveniently written as

$$^{232}\text{Th} + n \rightarrow {}^{233}\text{Th} \xrightarrow[22.1\ \text{m}]{\beta^-} {}^{233}\text{Pa} \xrightarrow[27.4\ \text{d}]{\beta^-} {}^{233}\text{U} \ . \qquad (2.33)$$

where we indicate the type of nuclear transformation and half-life for each of the processes.

Another important transmutation chain is the uranium-plutonium chain:

$$^{238}\text{U} + n \rightarrow {}^{239}\text{U} \xrightarrow[23.5\ \text{m}]{\beta^-} {}^{239}\text{Np} \xrightarrow[23.5\ \text{d}]{\beta^-} {}^{239}\text{Pu} \ . \qquad (2.34)$$

Both of these nuclear transmutations are shown in the form described on the Chart of the Nuclides, Fig. 2.8. We point out that the breeding chain may not proceed to its final stage if an additional nuclear transmutation occurs at any of the intermediate stages. For example, Uranium-239 could capture a neutron before it decays to Neptunium-239.

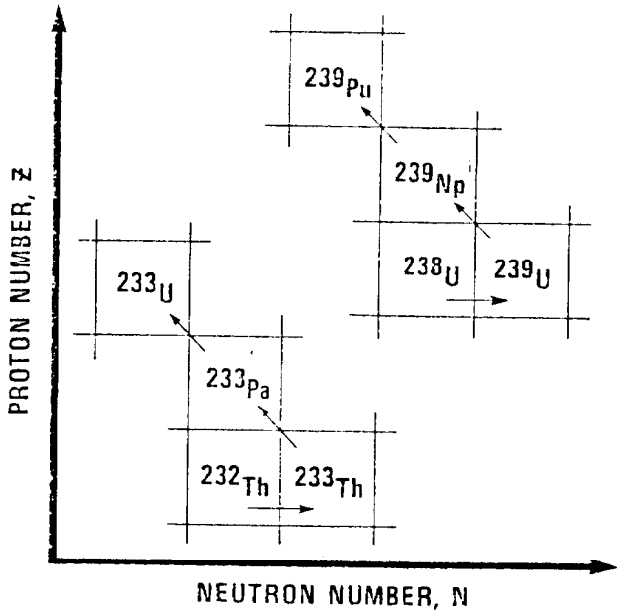

FIG. 2.8: Nuclear transformation associated with the production of fissile nuclei (Uranium-233 and Plutonium-239) from fertile nuclei (Thorium-232 and Uranium-238).

2.5 NUCLEAR FISSION PROCESS

Nuclear fission is the process whereby a nucleus breaks up into two relatively massive nuclear fragments. Although fission is known to occur spontaneously with some nuclei and may be induced by high energy reactions in others, the form of fission of primary interest in reactors is that induced by thermal neutrons in the three nuclei Uranium-233, Uranium-235, and Plutonium-239; note that all three nuclei are odd-numbered. Only Uranium-235 appears in nature to the extent of 0.72% is natural uranium; as indicated in the preceding section, the other nuclei can be produced by nuclear transmutation starting with Thorium-232 and Uranium-238. Less frequently occurring fission by high energy neutrons will be discussed at a later point.

A widely adopted model of the fission process is based on a liquid-drop analogy. The nucleus is assumed to possess an initial spherical shape based on a balance of surface tension forces and the electrostatic repulsion forces associated with the positively charged protons. The addition of a neutron to the nucleus adds to the excitation energy of the nucleus. The nucleus oscillates and, at various stages, takes on a dumb-bell like shape until it eventually breaks up. Several free neutrons and other forms of radiation are emitted simultaneously. This oscillatory sequence is illustrated in Fig. 2.9. An additional consequence of the fission process is the observation that the fission products are generally neutron rich and radioactive; as these nuclei decay with time according to the law of radioactive decay, Section 2.3, they emit neutrons which, as will become apparent later, are vital to the control of a nuclear reactor.

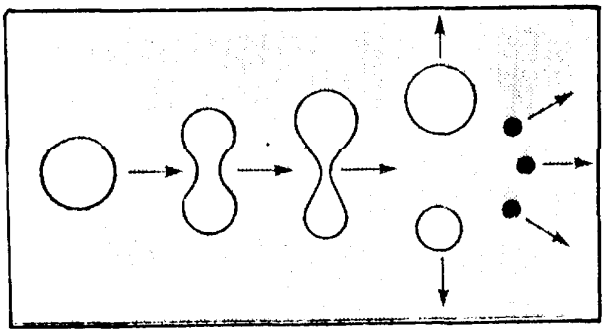

FIG. 2.9: Fission of a nucleus according to the liquid-drop model. The various stages are illustrated here proceeding from left to right; the high energy fission neutrons are represented by the black spheres.

The fission process is a statistical phenomena in the sense that the type of fission product, the number of fission neutrons, and other forms of radiation vary from one fission to the next but collectively represent a predictable

statistical distribution; indeed, the distribution of masses of fission products is known with considerable precision, Fig. 2.10. The variations among the three fissile nuclei are slight; also, the distributions exhibit a slight dependence on incident neutron energy.

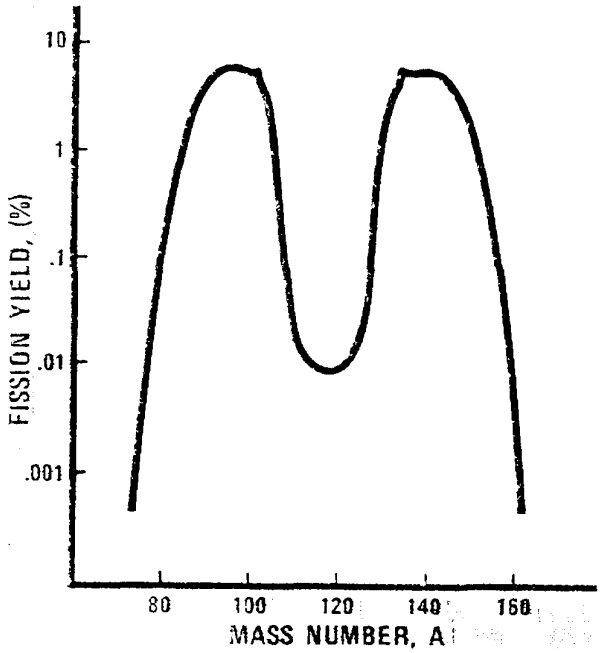

FIG. 2.10: Distribution of fission products for the various atomic mass numbers.

Of considerable consequence in nuclear reactor analysis is the number of fission neutrons released per neutron absorbed. Although this parameter is also a statistical variable in the sense that it varies from fission to fission, its dependence on incident neutron energy is the most important characteristic. This energy dependence is shown in Fig. 2.11. Note that it would not be desirable to have intermediate energy neutrons induce fission because the yield of fission neutrons is relatively low. Although the number of fission neutrons released increases dramatically if fission is induced by neutrons possessing an energy in excess of 10 MeV, we will show later that this is of little consequence because in an operating nuclear reactor there are very few neutrons with such a high energy. Thus, on the basis of these comments, it is clear that the variation in the number of fission neutrons emitted per fission, Fig. 2.11, favours a thermal nuclear reactor system. That is, a reactor designed so as to provide conditions which lead to most neutrons possessing energies in the thermal energy range, Fig. 2.11.

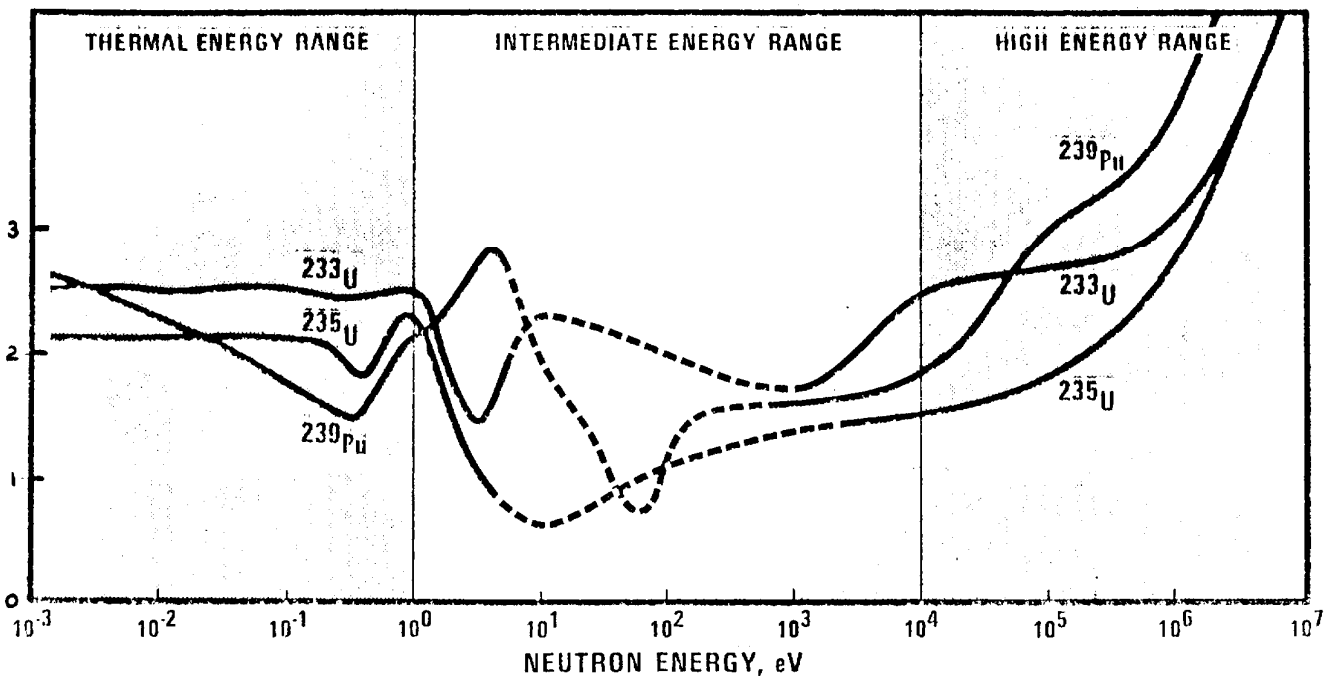

FIG. 2.11: Variation in the number of fission neutrons emitted as a function of incident neutron energy for three fissile nuclei.