# Chapter 1

# **Neutron-Nucleus Interactions and Fission**

The central problem in nuclear-reactor kinetics is to predict the evolution in time of the neutron population in a multiplying medium. Point kinetics allows the study of the global behaviour of the neutron population from the average properties of the medium. Before tackling, in the following chapters, the equations governing the time variation of the reactor power (proportional to the total neutron population), we shall discuss briefly the properties of a neutron-multiplying medium. After recalling a number of definitions, we shall give a qualitative description of the principal nuclear reactions at play in a self-sustaining chain reaction, and dwell on the source of fission neutrons. Since delayed neutrons play a crucial role in reactor kinetics, we shall describe in greater detail their production in a reactor.

#### **1.1** Neutron-Nucleus Interactions

A neutron is an elementary particle. It is stable when bound to a nucleus by the nuclear force. In its free state, a neutron decays, with a time constant of 12 minutes, to a proton, accompanied by the emission of a  $\beta$  particle and an antineutrino. In view of the fact that free neutrons in a reactor either are absorbed in matter or escape, on a time scale of less than 10<sup>-3</sup> s, it is clear that neutron instability is completely negligible in reactor physics.

The power generated in a reactor comes from neutron-induced fissions. It results from the interaction between the neutrons and the nuclei of the physical medium. It is generally possible to neglect neutron-neutron interactions, because the density of neutrons is much smaller than that of nuclei. Free neutrons have a certain speed relative to nuclei. If one neglects the energy of thermal motion of the nuclei, the kinetic energy E of a neutron represents the energy available to a nuclear reaction between it and a nucleus. If we denote the relative speed of a neutron by v and its mass by m1, then

$$E = \frac{1}{2}mv^2 \tag{1-1}$$

The energy is generally given in units of electron-volts ( $1 \text{ eV} = 1.6021 \times 10^{-19} \text{ J}$ ).

#### 1.1.1 Cross Sections

Interactions of neutrons with matter are described in terms of quantities called cross sections, which can be defined in the following fashion. Imagine a unidirectional neutron beam with a density of n neutrons per cm<sup>3</sup>, of speed v, impinging perpendicularly on a thin slice of a target with a density of N identical nuclei per cm<sup>3</sup>. Define the neutron flux  $\phi$  by

$$\phi = n v \tag{1-2}$$

<sup>&</sup>lt;sup>1</sup> The speed of neutrons in a reactor is sufficiently low that one can neglect relativistic effects. Therefore  $m = m_0$ , the neutron rest mass (1.6748 x 10<sup>-27</sup> kg).

Thus,  $\phi$  measures the number of neutrons crossing a unit surface area (1 cm<sup>2</sup>) per second. Experiments show that the rate R of interactions of neutrons per cm<sup>2</sup> of target area is proportional to the neutron flux, the density of nuclei, and the target thickness:

$$R = \sigma \phi N V \tag{1-3}$$

where the constant of proportionality  $\sigma$  is called the microscopic cross section. V is the volume of the target per cm<sup>2</sup> (i.e., the target thickness).

The microscopic cross section is a function of the speed of the neutrons relative to the nuclei of the target. It is therefore a function of the kinetic energy available to the reaction. One writes:

$$\sigma = \sigma(E) \tag{1-4}$$

Since NV is equal to the total number of nuclei per cm<sup>2</sup> of target area,  $\sigma$  is a measure of the probability that a neutron in the beam will have an interaction, per target nucleus. It is to be noted that  $\sigma$  has the dimensions of an area. Microscopic cross sections are usually measured in barns (b), where 1 b = 10<sup>-24</sup> cm<sup>2</sup>.

Neutrons can interact with nuclei in different ways; each type of interaction will be described in terms of a specific "partial" cross section. Since the probabilities are additive, the sum of the partial cross sections (the total cross section) is a measure of the probability of an interaction of any type when a neutron hits the target.

Let us now consider a physical medium with N nuclei per cm<sup>3</sup>. Let us imagine that there exists, within the medium, a density n of free neutrons per cm<sup>3</sup>, propagating at a speed v in all directions. The rate of interactions per cm<sup>3</sup> will then be

$$\frac{R}{V} = N \sigma n v \tag{1-5}$$
$$= \Sigma \phi$$

The constant of proportionality  $\Sigma$  between the rate of interactions and the scalar flux  $\phi$  is called the macroscopic cross section. Note that the microscopic cross section  $\sigma$  is a property of each type of nucleus for each type of interaction. If the physical medium contains many types of nuclei, it will be necessary to sum over all types in order to obtain the total macroscopic cross section:

$$\Sigma = \Sigma(E)$$

$$= \sum_{i} N_{i} \sigma_{i}(E)$$
(1-6)

The macroscopic cross section measures the probability of an interaction for a neutron travelling over a unit distance in the medium under consideration. The units of  $\Sigma$  are therefore cm<sup>-1</sup>.

Each type of interaction has its own particular cross section. The total cross section,  $\sigma_i$ , is the sum of the cross sections for scattering,  $\sigma_i$ , and for absorption,  $\sigma_a$ :

$$\sigma_t = \sigma_s + \sigma_a \tag{1-7}$$

#### 1. Neutron-Nucleus Interactions and Fission

The scattering cross section includes those for elastic scattering,  $\sigma_{e}$ , and for inelastic scattering,  $\sigma_{f}$ . In elastic scattering, the kinetic energy of the incident neutron is shared between the target nucleus and the neutron emerging from the collision. In an inelastic scattering, the kinetic energy is not conserved, the target nucleus being left in an excited state. One has:

$$\sigma_s = \sigma_e + \sigma_i \tag{1-8}$$

On the other hand, the absorption cross section includes those for fission,  $\sigma_{f}$  for radiative capture,  $\sigma_{v}$  and for reactions of the type (n,2n), (n,3n), etc.:

$$\sigma_a = \sigma_f + \sigma_\gamma + \sigma_{n,2n} + \sigma_{n,3n} + \cdots$$
(1-9)

The cross sections depend on the energy of the incident neutron, and this dependence is very complex. It results from the structure of the target nuclei and the type of interaction. The study of cross sections has been the concern of experimental and of theoretical nuclear physics for many years. Databases of standardised data have been created jointly by several national laboratories. These files (ENDF) contain an evaluation of all nuclear reactions for each nuclide, and include experimental analysis combined with the predictions of theoretical models, in a format allowing electronic processing of the data. Specialised computer programs, such as NJOY (MacFarlane, 1978) allow the preparation of data for neutronics calculations in the form of cross sections as functions of neutron energy. In this monograph, as in most treatises on neutronics, it is assumed that all cross sections  $\sigma(E)$  are available.

## **1.1.2** Types of Interaction

When a neutron comes into collision with a nucleus, three types of interaction are possible: formation of a compound nucleus, potential scattering, or direct interaction.

#### a) <u>Compound Nucleus</u>

In compound-nucleus formation, the incident neutron is absorbed by the target nucleus and a system called compound nucleus is formed. The neutron's kinetic energy is shared among all nucleons. If the target nucleus is  ${}_{Z}X^{A}$ , where Z is the number of protons, A the number of nucleons (also called the "mass number"), and X is the chemical symbol of the element, then the compound-nucleus formation can be represented as

$${}_{Z}X^{A} + {}_{0}n^{1} \rightarrow \left({}_{Z}X^{A+1}\right)^{*} \tag{1-10}$$

The energy of excitation (i.e., above the ground state) of the compound nucleus,  $E^*$ , is, however, larger than the incident neutron's kinetic energy (much larger if the incident neutron is in the thermal energy range). This is so for the following reason. The binding energy of a nucleon in a nucleus is the energy which must be supplied to the nucleon to free it from the nucleus. In the reverse process, this energy reappears when the neutron is captured into the nucleus<sup>2</sup>. Therefore, when the incident neutron is absorbed into the target nucleus to form the compound nucleus, it contributes its binding energy  $E_b$  to the compound nucleus. The energy of excitation of the

<sup>&</sup>lt;sup>2</sup> One can calculate the total binding energy of a nucleus from the mass defect,  $\Delta m$  (which is the difference between the mass of the bound nucleus and the sum of the masses of the nucleons), using Einstein's equation  $E_{Biot} = \Delta mc^2$ .

compound nucleus is consequently the sum of the incident neutron's kinetic energy and its binding energy in the compound nucleus:

$$\boldsymbol{E}^* = \boldsymbol{E} + \boldsymbol{E}_{\boldsymbol{b}} \tag{1-11}$$

The compound nucleus is highly unstable. It is in an excited state and lives only about  $10^{-14}$ Ês before de-excitation. This occurs by particle and/or photon emission. The type of nuclear reaction allowing the compound nucleus to reach stability is called a de-excitation pathway. The main de-excitation pathways of the compound nucleus are shown in Table 1.1.

The compound nucleus can also de-excite by re-emitting a neutron. If the residual nucleus  $_{Z}X^{A}$  is in its ground state, the process is called elastic scattering. More often the residual nucleus is left in an excited state (later de-exciting by emitting a photon), and the kinetic energy of the emitted neutron is smaller than that of the incident neutron: this is inelastic scattering.

Elastic resonant scattering (n, n)	$\left({}_{Z}X^{A+1}\right)^{*} \rightarrow {}_{Z}X^{A} + {}_{0}n^{1}$
Inelastic resonant scattering (n, n')	$\left(z^{X^{A+1}}\right)^* \to z^{X^A} + {}_0n^1 + \gamma$
Radiative capture (n, g)	$\left(z^{X^{A+1}}\right)^* \to z^{X^{A+1}} + \gamma$
Fission (n, f)	$(z^{X^{A+1}})^* \rightarrow c^{PF_1^B} + z_{-C}^{PF_2^{A+1-B-\nu}} + v_0^{n^1} + \gamma$
Transmutation (n, p)	$(z^{X^{A+1}})^* \rightarrow z_{-1}Y^A + {}_1H^1$
(n, a) reaction	$(z^{X^{A+1}})^{\bullet} \rightarrow z_{-2}Y^{A-3} + {}_{2}\mathrm{He}^{4}$
(n, 2n), (n, 3n) reactions	-

 Table 1.1
 De-excitation pathways for the compound nucleus

The compound nucleus can also de-excite to the ground state of the nuclide  ${}_{Z}X^{A+i}$  by emitting one or more  $\gamma$  rays. This is called radiative capture. In addition, depending on the random collisions between nucleons in the highly excited compound nucleus one or more nucleons (a proton, a deuteron, 2 or 3 neutrons, an  $\alpha$  particle, ...) can be ejected. These reactions generally require that the kinetic energy E supplied be above a minimum value. These are termed thresholdtype reactions.

In general, cross sections involving the capture or absorption of the neutron vary inversely with the neutron speed. This illustrates the fact that it is generally more difficult to hit a moving target than an stationary one. Also, according to quantum mechanics, a nucleus can exist only in certain states called quantum states. For a compound nucleus to be formed, it must be in an allowed intermediate state. If there is an excited state of the nucleus  ${}_{Z}X^{A+1}$  in the vicinity of  $E^* = E + E_b$ , the probability of compound-nucleus formation will be great, and the cross section for the reaction will be large. But if there is no allowed level close to  $E^*$ , the cross section will be much smaller. The dependence of the cross sections on the kinetic energy of the incident neutron will therefore exhibit resonances, which lead to sudden variations of the cross sections with neutron energy. The presence of resonances is illustrated in Figure 1.1.

## Fig. 1.1 Nuclear Cross section with Resonances



#### b) Potential Scattering

While the cross section for elastic scattering is significant only near a resonance, potential scattering is a type of elastic scattering which can take place for any energy of the incident neutron. This mode of interaction does not involve the formation of a compound nucleus. It appears simply as a result of the presence of the nucleus. It is a collision of the "billiard ball" type, in which the total energy is conserved. Potential scattering is a function only of the forces which act on the neutron in the vicinity of the target nucleus, and these forces depend on the dimensions and the shape of the nucleus.

If we assume the target nucleus to be at rest, energy conservation can be shown to imply that the kinetic energy E' of the neutron after the collision is always smaller than the incident energy E, and must be in the range

$$\alpha E \leq E \leq E \tag{1-12}$$

where

$$\alpha \approx \left(\frac{A-1}{A+1}\right)^2$$

The neutron loses therefore a fraction of its energy in each collision: we have neutron moderation (slowing down).

#### c) Direct Interaction

High-energy neutrons can also interact with nuclei by direct interaction. In this type of interaction, a direct collision between the neutron and the nucleus results in the ejection of one or more nucleons (protons or neutrons) and in the absorption of the incident neutron. However, very few neutrons in a reactor possess sufficient energy to enter into direct interaction. This type of interaction is thus of little interest in reactor physics.

# 1.1.3 Reactions of Importance in a Fission Reactor

Table 1.2 gives values of various cross sections for heavy nuclides which may be present in nuclear fuel. These cross sections are for thermal neutrons (E = 0.025 eV, v = 2200 m/s).

The nuclear reactions which are important in a fission reactor are the following

- Fission induced by neutrons absorbed by the fuel (much more probable for neutrons slowed to the thermal energy range). We will discuss this reaction in the next section. Spontaneous fission of uranium is also possible. However, this natural mode of disintegration has very low probability<sup>3</sup>, and can be completely neglected as soon as the reactor is operating, even at very low power.
- Neutron scattering (both elastic and inelastic). These reactions are responsible for the slowing down of neutrons in a thermal reactor, via collisions with the nuclei of the moderator and/or coolant. The degree of effectiveness of these reactions plays an important role in dictating the size (and therefore the cost) of the reactor. Indeed, neutrons emerging from fission in a nuclear reactor have an energy of the order of 2 MeV. The cross section for fission is, however, much larger at thermal energies (= 0.025 eV). It is therefore advantageous to introduce a moderator in the reactor; the intent is to slow the neutrons to thermal energies without absorbing them. The most effective moderator is the one having the smallest atomic mass (so that  $\alpha$  in equation 1-12 will be as small as possible) while having as small a capture cross section as possible. Table 1.3 shows the slowing-down power of various nuclides. It is clear from Table 1.3 why heavy-water (D<sub>2</sub>O) reactors are generally of larger size than light-water reactors.
- Radiative capture of neutrons in the fuel and the other materials in the reactor. This type of reaction is very important, because neutron economy is crucial for sustaining the chain reaction (radiative capture is a parasitic absorption when it does not lead to a fission). Transmutations in the fuel, resulting from radiative capture, play a very important role. For example, neutron capture in <sup>238</sup>U leads, following two  $\beta$  decays, to the production of <sup>239</sup>Pu. We note from Table 1.2 that the cross section of <sup>238</sup>U is relatively small for thermal neutrons. In spite of this, and in spite of the fact that there is no plutonium in fresh CANDU fuel, <sup>239</sup>Pu fissions represent about half of all fissions in a CANDU reactor. The high capture rate in <sup>238</sup>U is due to numerous resonances present at higher energies. On the other hand, <sup>240</sup>Pu has a very large capture cross section in the thermal range. Secondary nuclides (formed via neutron capture) are often very radioactive, which may result in a radiological concern in the operation of the reactor. For example, the production of tritium by radiative capture in deuterium is important in heavy-water reactors, such as CANDU. To be noted as well is the activation of corrosion products circulating in the heat-transport system (for example <sup>60</sup>Co<sub>277</sub>, <sup>56</sup>Mn<sub>25</sub>, and <sup>64</sup>Cu<sub>29</sub>).
- Radioactive decay of fission products. This plays a fundamental role in the design of a nuclear reactor, and has a direct effect on its behaviour, as we shall see throughout this book. We note that this radioactivity results in:
  - the emission of a large number of  $\gamma$  rays in the short term (leading to heat production, need for shielding, etc.),
  - the emission of  $\beta$  and  $\alpha$  radiation (leading to the long-term radioactivity of spent fuel), and
  - the emission of neutrons (delayed relative to fission).

Other reactions, such as (n,2n) reactions, are less important, because the reaction threshold is higher than the energy of most of the neutrons in the reactor (the threshold values are given in Table 1.4). One must still take them into account if one wishes to know accurately the composition of irradiated fuel (from the point of view of waste storage). Finally, we mention reactions of the type  $(\gamma,n)$  and  $(\gamma,fission)$ , which are generally not important for reactor operation, except for  $(\gamma,n)$ reactions in heavy-water reactors (section 1.3.3).

<sup>&</sup>lt;sup>3</sup> For example, the half-life of <sup>238</sup>U for  $\alpha$ -decay is 4.5x10<sup>-9</sup> years, and for spontaneous fission it is 5x10<sup>5</sup> years. The rate of disintegration by spontaneous fission corresponds to 10<sup>-12</sup> % of full power in a CANDU reactor.

Noyau	σ <sub>f</sub>	σg	σa
U233	525.0	53.0	578.0
U <sup>235</sup>	577.0	106.0	683.0
U238	-	2.70	2.70
Pu <sup>239</sup>	742.1	286.9	1029.1
Pu <sup>240</sup>	-	277.9	277.9
Pu <sup>241</sup>	1015.2	381.0	1396.2

Table 1.2Thermal Neutron Cross section (0,025 eV), in barns<br/>(Westcott, 1960)

Table	1.3	Neutron	Slowing	Down	Properties
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Isotope	<u>A</u>	<u>a</u>	N
Hydrogen (H <sup>1</sup> )	1	0	18
Deuterium (H <sup>2</sup> )	2	0.111	25
Graphite (C <sup>12</sup> )	12	0.716	115
Uranium (U <sup>238</sup> )	238	0.983	2172

N = number of collisions required to slow down a neutron from 2 MeV to 0.025 eV via potential scattering. For large  $A, N \approx 9A + 6$  (Weinberg, 1958).

Novau	(n 2n)	(n 3n)
1107au		
U <sup>233</sup>	5.9	13.2
U <sup>235</sup>	5.2	12.0
U <sup>238</sup>	6.1	11.5
Pu <sup>239</sup>	5.5	12.5

# Table 1.4 Reaction Threshold, in MeV(Lamarsh, 1966)

#### 1.2 Fission

Fission is obviously the most important reaction in a reactor. In this section, we discuss briefly the characteristics of energy production by fission, as well as the distribution of fission products.

# 1.2.1 Energy Produced in Fission

Let us consider, as an example, a fission induced by the absorption of a neutron in  $^{235}$ U:

$${}_{92}U^{235} + \underbrace{0^{n^{1}}}_{\text{Thermal Neutron}} \rightarrow \underbrace{(}_{92}U^{236} \underbrace{)^{r}}_{\text{Fission Products}} \rightarrow \underbrace{}_{36}\underbrace{\text{Kr}^{95} + }_{56}\underline{\text{Ba}^{139}}_{\text{Fission Products}} + \underbrace{2}_{0}\underbrace{0^{n^{1}}}_{\text{Fast Neutrons}} + \gamma$$

The largest fraction of the energy released in fission appears as the kinetic energy of the fission fragments (<sup>95</sup>Kr and <sup>135</sup>Ba in the above example). The remainder is shared among the particles emitted in fission, as shown in Table 1.5.

It is important to distinguish between the total energy produced in fission and the energy recovered in a reactor. The fission fragments are completely ionized and quickly slowed down, as any charged particle moving in a solid. The kinetic energy of the fission fragments is therefore quickly absorbed within a few micrometers of the site of the fission.

The fission products are generally unstable. They decay, emitting  $\beta$  and  $\gamma$  radiation and neutrinos. The  $\beta$  rays have a short range and are absorbed within the fuel, while the  $\gamma$  radiation is absorbed in the reactor or in the shields. The neutrinos, on the other hand, escape completely. About 5 MeV of the kinetic energy of the prompt neutrons emitted in fission are recovered in the moderator. Of the v (2 or 3, on the average 2.4) neutrons emitted, only one needs to be absorbed in the fuel to sustain the chain reaction. The remaining (v-1) neutrons will therefore be absorbed in parasitic capture. Radiative capture of these extra neutrons will produce from 3 to 12 MeV of  $\gamma$ radiation, depending on the reactor materials. We note that the capture- $\gamma$  energy compensates approximately for the energy lost in the escape of the neutrinos.

The energy released during or after fission is about 200 MeV ( $3.2 \times 10^{-11}$  J) per fission.. It varies slightly from one fissionable isotope to another (it is approximately 4% higher in <sup>239</sup>Pu). Thus, the complete fission of one gram of <sup>235</sup>U produces 2.28 x 10<sup>4</sup> kW.h/g, i.e., 0.95 MW.d/g.

Form	Energy Released	Useful Energy
	(MeV)	(MeV)
Fission fragments	168.2	168
Fission products:		
β	7.8	8
γ̈́	6.8	7
neutrinos	≈ 12	
Prompt ys	7.5	7
Prompt neutrons	4.8	5
Radiative-capture	-	3-12
γs		`
Total	≈ 207	198-207

Table 1	.5	Energy	Released	рег	Fission	in	<sup>235</sup> U	(Keepin	, 1965	)
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It is clear that the mass of fuel required to operate a nuclear generating station is relatively small. For instance, a nuclear reactor producing 1000 MW(e) [i.e.,  $\approx$  3000 MW(th)] consumes about 1150 kg of fissile material per year.



Fig. 1.2 Average Binding Energy per Nucleon

One can understand why energy is released in fission by considering that the components of the nucleus are bound together by strong nuclear forces, more powerful than the Coulomb repulsion between the protons. When fission occurs, these forces are released as in a spring, and the fission fragments are ejected with a large amount of kinetic energy, corresponding to the binding energy of the nucleons.

The average binding energy per nucleon varies from one nuclide to another, as illustrated in Figure 1.2. It increases with mass number for the light nuclides, reaches a maximum in iron, and decreases slowly thereafter for the heavy nuclides. Thus, a net energy gain results from the fission of heavy elements (or from the fusion of light elements). A more stable configuration results when a heavy nuclide breaks up into two lighter nuclides: this is the reason for the existence of fission. One could therefore expect that all nuclei with A > 50 would fission spontaneously, which is of course not the case.

For fission to occur, energy must be supplied to the nucleus in some way. In other words, a potential barrier acts against fission. One can understand the phenomenon of fission intuitively with the help of the droplet model. In general, nuclei do not fission, for the same reason a drop of water does not break up into two fragments, even if the energy balance is favourable. In the case of the drop, the surface tension acts as a barrier which tends to keep the fragments together (Figure 1.3).

In the case of the fissioning nucleus, it is the attractive force between nucleons which holds the fragments together. At the start, the potential energy of the nucleus is equal to the binding energy of the nucleons (the kinetic energy of the fragments is zero). In order to deform the nucleus, energy must be supplied to increase the average separation distance between nucleons. The potential energy of the nucleus increases. The nuclear force is, however, of short range. As separation of the fragments proceeds, the repulsive force between them diminishes, leading to a decrease in the potential energy as s increases. As a result of these opposing effects, the potential energy exhibits a maximum (of approximately 6 MeV) at some separation; this peak constitutes the

barrier to fission. In spite of the barrier, quantum mechanics allows spontaneous fission to occur (though with very low probability) via a "tunnelling effect", i.e., without energy input.

As indicated earlier, in any reaction involving a neutron capture and the formation of a compound nucleus, the energy supplied to the nucleus comes from the binding energy  $E_b$  of the incident neutron (in the compound nucleus) and its kinetic energy E. If the binding energy of the incident neutron is higher than the barrier to fission, the nucleus is fissile, i.e., it can undergo fission induced by thermal neutrons. On the other hand, if this energy is insufficient and it is necessary to add the neutron's kinetic energy to surmount the fission barrier, the nucleus is not fissile, but is fissionable. [Note: fissile nuclides are also fissionable.] Most heavy nuclides beyond <sup>232</sup>Th are fissionable. The only naturally occurring fissile nuclide is <sup>235</sup>U, which is present in the proportion of 0.71% in natural uranium. The other fissile nuclides are produced artificially in reactors.

Fig. 1.3 The Droplet Model



One can understand why some nuclides are fissile and others are not in the following way. In a nucleus the protons tend to pair up, as do the neutrons. Consequently, the binding energy of a neutron absorbed into a nucleus containing initially an odd number of neutrons will generally be higher. It will be easier to surmount the fission barrier in such a case. One does indeed find that nuclides with even Z and odd A are easier to fission than those with even Z and even A.

In general, if Z is even and A is odd, the nuclide can undergo fission induced by both fast and thermal neutrons: thus <sup>233</sup>U, <sup>235</sup>U, <sup>239</sup>Pu, and <sup>241</sup>Pu are fissile nuclides. One can also see from Table 1.2 that the cross section for fission increases with the mass number A.

On the other hand, if A is even, fission can be induced by fast neutrons only. Thus, <sup>232</sup>Th, <sup>238</sup>U, and <sup>240</sup>Pu are fissionable nuclides. They are also "fertile" nuclides, because neutron capture in them leads to the production of fissile nuclides (<sup>235</sup>U, <sup>239</sup>Pu, and <sup>241</sup>Pu).

#### **1.2.2** Distribution of Fission Products

In general, only two fission fragments are observed, i.e., fission is binary. The kinetic energy of the fission fragments is greater than 80% of the energy from fission. Isotopes of more than thirty elements are observed among fission products. The yield of a fission product is defined as the number of nuclei of that element obtained, on the average, per fission. It is observed experimentally that the mass of the fission fragments is in general not uniform. It is much more probable to find two fragments of unequal mass than two fragments of equal mass. In fact, as illustrated in Figure 1.4, the most probable ratio of fragment masses is about 1.46 (for 235U, the most probable masses are 95 and 139 [Bussac, 1978]).

It is also to be noted that most of the fragments or products of fission are unstable. The region of nuclide stability in the N vs. Z diagram (number of neutrons versus number of protons) is shown in Figure 1.5. The line of stability is not a straight line because, as Z increases, it is necessary to have a greater and greater number of neutrons to hold the nucleus together and surmount the Coulomb repulsion, which increases with the number of protons. The fission products often exhibit, therefore, an excess of neutrons relative to the number of protons. This excess is "corrected" by means of successive  $\beta$ - decays (which transform a neutron in the nucleus into a proton, with the emission of an electron). Sometimes, however, fission products reach stability by emitting neutrons. As we shall see later, this latter property is extremely important in reactor kinetics.



Fig. 1.4 Fission Product Yield for U<sup>235</sup>

Fig. 1.5 The Region of Nuclide Stability



#### **1.3 Production of Neutrons**

One of the most important consequences of fission is the emission of neutrons, leading to the possibility of a chain reaction. The great majority of the neutrons are emitted essentially instantaneously, within  $10^{-17}$  s. These are the *prompt neutrons*. A very small fraction of the neutrons from fission appears much later; this constitutes the *delayed neutrons*. The number of prompt neutrons emitted varies from one fission to another, ranging from 0 to 5. The *average* number of neutrons emitted per fission is needed for the purpose of calculations. This quantity, generally represented by the symbol v, includes the delayed as well as the prompt neutrons:

$$v = v_p + v_d \tag{1-13}$$

It is found experimentally that the average value of v depends on the fissioning nuclide and on the energy of the incident neutron inducing the fission. It is observed that v increases almost linearly with the incident neutron's energy E, over a wide range of E:

$$v(E) = v_o + aE \tag{1-14}$$

Experimental values of  $v_0$  are given in Table 1.6. Note that for <sup>238</sup>U the threshold for fastneutron fission limits the domain to  $E \ge 1.2$  MeV. When v(E) is averaged over the fission-neutron spectrum (see further below), an average value of 2.84 is found for <sup>238</sup>U. It is also to be noted that the average neutron yield in fission is in general higher for plutonium than for uranium.

Table 1.6	Average	Number of	Fission	Neutrons	(equation	1-14)
	•	(Keepin,	1965)			

Isotope	٧ <sub>0</sub>	a	E Domain
			(MeV)
<sup>233</sup> U	2.482	0.075	$0 \leq E \leq 1$
	2.410	0.136	E > 1
<sup>235</sup> U	2.432	0.065	$0 \leq E \leq 1$
	2.349	0.150	E > 1
<sup>238</sup> U	2.304	0.160	$E \ge 1.2$
-			(threshold)
<sup>239</sup> Pu	2.867	0.148	$0 \le E \le 1$
	2.907	0.133	E > 1

#### **1.3.1** Production of Prompt Neutrons

During fission, the fission fragments are created in an excited state. One or more prompt neutrons will be emitted when the excitation energy  $E^*$  is greater than the binding energy  $E_b$  of a neutron in the fragments. When this is the case, there is competition between the two possible deexcitation paths: emission of one or more  $\gamma$  rays or emission of a neutron. The process is illustrated in Figure 1.6.

Since the excitation energy is variable, there will therefore be a continuous distribution of energy of both the  $\gamma$  rays and the prompt neutrons. Both will have a characteristic energy spectrum. Given the dependence of the cross sections on the energy of the neutrons in the reactor, it will be important to know accurately the energy distribution of fission neutrons. In particular, the prompt neutron spectrum,  $\chi_p(E)$ , is defined in such a way that  $\chi_p(E)dE$  is the number of neutrons emitted with an energy between E and E + dE, per fission neutron; i.e.,  $\chi_p(E)$  is normalized in such a way that

$$\int_0^\infty \chi_p(E) \, dE = 1 \tag{1-15}$$

An analytic expression which reproduces experiment quite well for <sup>235</sup>U is (Lamarsh, 1966):

$$\chi_p(E) = 0.453 e^{-1.036E} \sinh \sqrt{2.29E}$$
 (1-16)

This relation is sketched in Figure 1.7. It characterises fairly well the prompt-neutron spectrum for all fissionable nuclides. The average energy of prompt neutrons can be found by integrating over the spectrum. We find:

$$\overline{E} = \int_0^\infty \chi_p(E) E \, dE = 1.98 \text{ MeV}$$
(1-17)

The most probable energy of prompt neutrons corresponds to the peak of Figure 1.7; the value is approximately 0.85 MeV.

#### **1.3.2** Production of Delayed Neutrons

The majority of fission products are unstable, and a large number of them are  $\beta$ - emitters. Some of them are precursors of delayed neutrons, in the sense that the nuclide resulting from the  $\beta$ decay is a neutron emitter, the neutron emission being delayed relative to the fission. Figure 1.8 illustrates the mechanism of emission of delayed neutrons.



Fig. 1.6 Prompt Neutron Emission

Fig. 1.7 The Prompt Neutron Spectrum (Eq. 1-16)



#### 1. Neutron-Nucleus Interactions and Fission

Suppose that nuclide  ${}^{A}X_{Z-J}$  is one of the fission fragments, and that it is a  $\beta^{-}$  emitter. A certain time after fission (characterized by the decay constant  $\lambda$  of the fission product), a  $\beta^{-}$  particle (electron) is emitted, simultaneously with a neutrino. The latter shares with the electron the energy available from the reaction (continuous energy spectrum for the emitted  $\beta^{-}$ ). Depending on the energy removed by the  $\beta^{-}$ , the resulting nuclide  ${}^{A}X_{Z}$  will have a larger or smaller excitation energy. If the  $\beta^{-}$  has the maximum available energy, which we will label  $E_{\beta}^{max}$ , the resulting nuclide will be in its ground state.

The emission of a delayed neutron by  ${}^{A}X_{z}$  will be possible only if the excitation energy  $E^{*}$  of the latter is greater than the binding energy  $E_{b}$  of the neutron. Consequently  ${}^{A}X_{z-1}$  will be a delayed-neutron precursor only if  $E_{\beta}^{max}$  is greater than the binding energy  $E_{b}$  of the neutron in  ${}^{A}X_{z}$ , that is, only if

$$E_{\beta}^{max} \ge E_b \tag{1-18}$$

From this it is clear that relatively few fission products are delayed-neutron precursors.

As the neutron is emitted within  $10^{-14}$  s, the *delay* between fission and the emission of the delayed neutron is due entirely to the lifetime for  $\beta^-$  decay (inverse of the decay constant  $\lambda$ ). In addition, since the distribution of fission products varies from one fissionable nuclide to another, each of the latter will have a characteristic set of delayed-neutron precursors. Consequently, for each fissionable nuclide, a certain number of delayed neutrons  $(v_a)$  will be emitted, on the average, per fission. This number is added to  $v_a$  in equation (1-13).

The *delayed-neutron fraction* is defined by the ratio

$$\beta = \frac{v_d}{v} \tag{1-19}$$

It is experimentally verified that the delayed-neutron fraction is in general very small. It is typically smaller than 1%. As in the case of the fission-neutron yield, the average number of delayed neutrons is a function of the incident neutron inducing the fission, although the dependence of  $v_d$  on E is lesser. Tuttle (1975) reports a 3% increase in the value of  $v_d$  for fast-neutron fission in <sup>235</sup>U. This variation results from the effect of E on the yield of fission products, some of which are delayed-neutron precursors (Figure 1.4).

Experimental values of the average delayed-neutron yield are given in Table 1.7 for thermal fission. It can be seen that there is still some uncertainty in the values of  $v_d$  for the principal nuclides. The percent values shown reflect the variation in the various experimental values reported.

Nuclide	V <sub>d</sub>	β
233U	0.00667	0.0027
<sup>235</sup> U	0.0166 ± 3 %	0.0068
<sup>239</sup> Pu	0.00654 ± 4 %	0.0023
<sup>241</sup> Pu	0.0154	0.0052
<sup>238</sup> U*	0.043 - 0.047	0.0158

Table 1.7Average Delayed-Neutron Yields in Thermal Fission<br/>(Blachot, 1990)

\* Average over fission-neutron spectrum



#### 1. Neutron-Nucleus Interactions and Fission

#### **1.3.3** Photoneutrons

Neutrons can be produced in a reactor by reactions of the type  $(\gamma,n)$ , but only if the  $\gamma$  has sufficient energy, because it is a reaction with a threshold. Such a reaction is observed in deuterium and beryllium. This reaction is therefore very important in heavy-water reactors, and more particularly in CANDU reactors, where more than 90% of the volume is occupied by the heavy-water (D<sub>2</sub>O) moderator and coolant. The reaction which leads to the production of photoneutrons (for  $\gamma$  rays of energy  $E_{\gamma} \ge 2.2$  MeV):

$$H^2 + \gamma \rightarrow H^1 + {}_0n^1 + \gamma' \tag{1-20}$$

One must distinguish between prompt  $\gamma$  rays (emitted in fission) and  $\gamma$  rays emitted in fissionproduct decay. Prompt photoneutrons are only marginal, because only a fraction of the  $\gamma$ 's have sufficient energy, and the number of prompt photoneutrons is much smaller than the number of prompt neutrons produced in the fuel in fission. On the other hand, photoneutrons produced by fission-product  $\gamma$ 's are *delayed neutrons* in their own right, since their production must await the decay of the fission products. Since the photoneutron precursors are not generally the same as the fuel delayed-neutron precursors, photoneutron production will be characterized by different decay constants  $\lambda$  and yields  $v_{d}$ . Indeed, some photoneutron precursors have a lifetime much longer than that of most delayed-neutron precursors (for example, 15 days versus 1 minute).

Comparative studies (with and without heavy water) have shown that the photoneutron yield varies as the delayed-neutron yield from one fissionable nuclide to another. The photoneutron yield is approximately 5% of the total delayed-neutron yield.

#### **1.4 Delayed-Neutron Groups**

Knowing the delayed-neutron yield  $(v_d)$  of each fissionable nuclide is not sufficient for kinetics calculations; the delay in the emission of the delayed neutrons must also be well known. The time behaviour of delayed-neutron emission can be obtained in two ways.

First, the classical approach (Keepin, 1965), which consists of measuring directly the source of delayed neutrons following a fission pulse in a sample of material. These macroscopic results are then analyzed by curve fitting into a certain number of delayed groups (generally 6), each group having its characteristic time constant and yield.

The second approach is more microscopic. Since in fact the delayed-neutron source is the decay of individual fission products, one can in principle derive the characteristics of the delayed-neutron emitters from the knowledge of the individual fission-product yields and the emission probabilities. In the last 20 years, advances in nuclear physics in the area of the detailed structure of nuclei, as well as progress in experimental techniques for isotopic separation and neutron spectroscopy, have contributed to improve the data base of individual yields and of delayed-neutron spectra from various fissionable nuclides. While the half-life and abundance of several precursors are not known precisely, the use of models has made it possible to extend the data bases to 271 precursors. This data has allowed the derivation of 6-group macroscopic data for 43 fissionable systems, ranging from <sup>227</sup>Th to <sup>255</sup>Fm (Brady, 1989). These macroscopic results, derived from microscopic data, compare well with direct measurements.

While it is possible, an individual treatment of the precursors is not practical for reactor-physics calculations. One uses instead the macroscopic data (measured or derived) to characterize the totality of the fission-product delayed-neutron precursors of a given fissionable nuclide.

# 1.4.1 Characterization of Delayed-Neutron Groups

Let us consider as an example the procedure used to obtain the properties of each of the precursor groups (macroscopic data).

One subjects a sample of material (fissionable nuclide *i*) to a *pulse* of neutrons. This neutron flux causes a certain number of fissions in the sample,  $n_{fi}$ , and the production of a set of precursors,  $v_{a}n_{fi}$ . The decay of these precursors gives rise to a decreasing source of (delayed) neutrons, which can be measured. An example of the decreasing source is sketched in Figure 1.9.



Fig. 1.9 The Delayed Neutron Source

The evolution of the count rate reflects the decay of all the fission products in the sample which are delayed-neutron precursors. One can fit this curve with a limited number of exponentials. A 6-term fit is usually performed. For each fissionable nuclide i, a relation of the following type is obtained:

$$S_{di}(t) = n_{fi} \sum_{k=1}^{6} v_{dki} \lambda_{ki} e^{-\lambda_{ki}t}$$
(1-21)

If several nuclides contribute to the delayed-neutron source, one should sum:

$$S_d(t) = \sum_{i} \left[ n_{fi} \sum_{k=1}^{6} v_{dki} \lambda_{ki} e^{-\lambda_{ki}t} \right]$$
(1-22)

which implies the knowledge of the decay constants  $\lambda_{ki}$ , for each family or group k of precursors and each nuclide i. One generally finds in references 6 group decay constants  $\lambda_k$  and 6 values of  $v_{dk}$  for each fissionable nuclide (Keepin, 1965; Brady, 1989).

#### 1. Neutron-Nucleus Interactions and Fission

It is remarkable that the decay constants of the precursor groups are almost the same from one nuclide to another. It is in fact understandable, since the same precursors are often found as fission products of the various nuclides. For instance <sup>87</sup>Br (Figure 1.9), with a half-life of 54.5 s, is a fission product of high yield from most fissionable isotopes. The existence of this fission product influences significantly the decay constant of the first delayed-neutron group for most fissionable nuclides.

This similarity between precursors for the different fissionable nuclides suggests a simplified approach to the treatment of the delayed-neutron groups (Calahan, 1973). The idea is to choose a single set of decay constants for all fissionable isotopes ( $\lambda_k$ , k = 1, 6), and redo the fit described above, obtaining new values of  $v_{di}$ . The source (equation 1-21) now becomes

$$S_{di}(t) = n_{fi} \sum_{k} v_{dki} \lambda_{k} e^{-\lambda_{k} t}$$
(1-23)

This is the approach selected in this monograph, following the lead of Ott (1985). The decay constants in Table 1.8 will therefore be used in all our numerical examples.

#### **1.4.2 Delayed-Neutron Fraction**

The set of delayed-neutron precursors for each of the main fissionable isotopes in the fuel will therefore be characterized by the 6 values of the yield  $v_{dki}$ , for the 6 groups k (Ott, 1985). Dividing the  $v_{dki}$  by the total yield  $v_i$ , as per equation 1-19, one obtains the delayed-neutron fractions  $\beta_{ki}$  in Table 1.9 for the 6 groups k. These values correspond to the  $\lambda_k$  values in Table 1.8; they will be used for our numerical results in the following chapters.

Note that the total delayed-neutron fraction for nuclide *i* is simply

$$\beta_i = \sum_k \beta_{ki} \tag{1-24}$$

The values in Table 1.9 do not include the photoneutrons, produced in the heavy-water moderator of CANDU reactors by  $(\gamma,n)$  reactions. The values for the *additional 9 groups* of delayed neutrons have been assembled by Kugler (1975) to represent the effect of the photoneutron source in the heavy-water moderator. These values are shown in Table 1.10.

The importance of the photoneutrons relative to that of the delayed neutrons from the fuel is apparent when one considers the evolution of the delayed-neutron source in a CANDU reactor following a shutdown. The following source is sketched in Figure 1.10 for a reactor shut down from full power (100% FP):

$$S_d(t) = 100 \% \cdot \sum_{k=1}^{15} \beta_k e^{-\lambda_k t}$$
(1-25)

The inclusion of the 9 photoneutron groups in the delayed-neutron source is obviously very important in the kinetics of heavy-water reactors.

Group	$\lambda$ (s <sup>-1</sup> )	Half-life (s)
1	0.0129	53.73
2	0.0311	22.29
3	0.134	5.17
4	0.331	2.09
5	1.26	0.55
6	3.21	0.22

Table 1.8Decay Constants for the Delayed-Neutron Groups,  $\lambda_k$ (Ott, 1985)

Table 1.9 Fraction of Delayed Neutrons from the Fuel,  $\beta_{ki}$  (%)

Group	<sup>235</sup> U	<sup>239</sup> Pu	<sup>241</sup> Pu	<sup>238</sup> U
1	0.0251	0.0087	0.0066	0.0206
2	0.1545	0.0639	0.1234	0.2174
3	0.1476	0.0493	0.0932	0.2570
4	0.2663	0.0747	0.2102	0.6156
5	0.0756	0.0235	0.0981	0.3570
6	0.0293	0.0080	0.0086	0.1190
Total β	0.6984	0.2281	0.5389	_ 1.587

Table 1.10Fraction of Delayed Neutrons from the Moderator<br/>(Photoneutrons),  $\beta_k(\%)$  (Kugler, 1975)

Group	T <sub>1/2</sub>	$\lambda_{k}(s^{-1})$	β <sub>k</sub> (%)
7	12.8 d	6.26 x 10 <sup>-7</sup>	1.653 x 10 <sup>-5</sup>
8	2.21 d	3.63 x 10 <sup>-6</sup>	3.372 x 10 <sup>-5</sup>
9	4.41 h	4.37 x 10 <sup>-5</sup>	1.058 x 10 <sup>-4</sup>
10	1.65 h	1.17 x 10 <sup>-4</sup>	7.670 x 10 <sup>-4</sup>
11	27 min	4.28 x 10 <sup>-4</sup>	6.779 x 10 <sup>-4</sup>
12	7.7 min	1.50 x 10 <sup>-3</sup>	1.101 x 10 <sup>-3</sup>
13	2.4 min	4.81 x 10 <sup>-3</sup>	2.298 x 10 <sup>-3</sup>
14	41 s	1.69 x 10 <sup>-2</sup>	6.694 x 10 <sup>-3</sup>
15	2.5 s	2.77 x 10 <sup>-1</sup>	2.136 x 10 <sup>-2</sup>
TOTAL (photoneutrons)	16.7min*	6.92 x 10 <sup>-4</sup>	3.306 x 10 <sup>-2</sup>

\* Average values:  $\lambda_{pk} = \sum (\beta_k / \lambda_k) / \sum \beta_k$  and  $T_{1/2} = \ln 2 \cdot \lambda_{pk}$ 



Fig. 1.10 Delayed Neutron Source in a CANDU (after shutdown from 100%FP)

#### 1.4.3 Delayed-Neutron Emission Spectrum

We have seen that the average number of delayed neutrons emitted per fission,  $v_d$ , varies from one fissionable isotope to another and that it depends slightly on the energy of the fission-inducing incident neutron.

The energy of delayed neutrons is generally much smaller than that of prompt neutrons: the average energy of delayed neutrons ranges between 465 keV for group 1 (the long-lived precursors) and 505 keV for group 6 (short lifetime), versus 2 MeV for prompt neutrons (Brady 1989).

By analogy with the prompt-neutron spectrum, one defines a *delayed-neutron spectrum* to describe the energy distribution of delayed neutrons at birth. Since this distribution depends on the energy of excitation of the emitters, the emission spectrum will depend on the delayed-neutron family (group). There will thus be a different spectrum for each precursor group and each fissionable isotope. The determination of the detailed spectra is the object of nuclear-physics research, and the data bases are continually updated to include more recent data (for example ENDF/B-VI). The spectrum  $\chi_{ak}(E)$  is defined in such a way that  $\chi_{dk}(E)dE$  is the number of delayed neutrons emitted with an energy ranging between E and E + dE, for each of the  $v_{dk}$  delayed neutrons emitted by the precursors of group k. The spectra are therefore normalized as in equation 1-15. Figure 1-11 shows a delayed-neutron spectrum from ENDF/B-I (obsolete).

Very detailed measurements have tried to establish, without success, the influence of the energy of the fission-inducing neutron on the delayed-neutron spectrum (Sharfuddin, 1988). It has therefore been effectively observed that the delayed-neutron spectrum is *independent of the neutron's incident energy*.

If one introduces an energy dependence into delayed-neutron emission in equation 1-23, one can write the energy-dependent delayed-neutron source from a fission impulse in a sample containing only one fissionable nuclide as

$$S_d(E,t) = n_f \sum_k \chi_{dk}(E) \, v_{dk} \, \lambda_k e^{-\lambda_k t} \tag{1-26}$$

Since each precursor group has its own decay constant, the total delayed-neutron spectrum varies with time during a transient. In order to illustrate this variation, let us consider the following three extreme cases:

• the initial spectrum:

$$S_d(E,0) = \sum_k \chi_{dk}(E) \, v_{dk} \, \lambda_k \tag{1-27}$$

• the steady-state emission spectrum, equivalent to integrating  $S_d$  over all time:

$$S_d^{stat}(E) = \int_0^\infty S_d(E, t) dt \propto \sum_k \chi_{dk}(E) v_{dk}$$
(1-28)

• The asymptotic spectrum, which dominates the long-term source and corresponds essentially to the long-lived precursor group (smallest value of  $\lambda_k$ , k=1):

$$S_d^{as}(E) \propto \chi_{d1}(E)$$
 (1-29)

Comparing these extreme cases shows that the total spectrum of delayed neutrons in a reactor changes with time. The delayed-neutron spectrum in steady-state reactor operation is therefore generally different from that in transient states.

## 1.5 Conclusion

The concept of a fission reactor is based on the principle of the chain reaction, with fission of heavy nuclei in the fuel resulting from the absorption of neutrons born in previous fissions. The energy released in fission is deposited in the fuel. In power reactors, this energy is removed by a coolant and delivered to a steam generator (SG) and then to a turbine generator for the production of electricity.

The only fissile isotope present in nature is <sup>235</sup>U, found in natural uranium in a proportion smaller than 1%. So, most operating reactors in the world utilise a uranium fuel cycle.

One must distinguish fast-neutron ("fast") reactors from thermal reactors. In thermal reactors, one makes use of the fact that fission cross sections are larger for slow neutrons than for fast

neutrons: a moderator is introduced in the reactor to slow neutrons down before they are absorbed in the fuel. The moderator most often used is light water, which has the advantage of low cost and requires a relatively small volume. It is this type of reactor (LWR) which is used in naval propulsion.

The relatively large absorption of neutrons in light water requires however the use of fuel enriched in the fissile isotope (<sup>215</sup>U), which increases the operating cost. The use of heavy water as moderator eliminates this problem, and allows the reactor to operate on natural-uranium fuel (CANDU). Capital costs are higher however. One can also use graphite as the moderator, particularly in reactors cooled with gas (e.g., helium), as graphite can withstand very high temperatures (HTGR).

The neutron flux will generally be higher in fast reactors (for the same power density), because the cross sections are smaller. One uses a liquid metal (sodium) as heat-transport fluid, which allows very high temperatures at low pressures (LMFBR). The much harder neutron spectrum also allows taking advantage of neutron capture in <sup>238</sup>U to attain a breeding cycle, with more fissile material produced (as <sup>239</sup>Pu) than is destroyed.

We shall not treat here the engineering aspects of nuclear reactors (see for example Glasstone, 1982). The theory covered in the following chapters is, however, applicable to all types of fission reactors, and will allow the reader to understand their transient behaviour.



• • • •

Fig. 1.11 Delayed Neutron Emission Spectrum (Ott, 1985)