CHAPTER 4

NEUTRON SOURCES

I. The Nuclear Reactor

(A) Fission

The operation of a nuclear reactor is based on the fission reaction. In 1939 O. Hahn and F. Strassmann discovered that a uranium nucleus, after absorbing a thermal neutron, could split into two new nuclei. Each of those fragments, called fission products, carries *about* half the weight and half the charge of the uranium nucleus, and possesses an extremely high kinetic energy.

The principle of mass and energy conservation allows the calculation of the energy, released by the hypothetical fission of a ^{235}U nucleus. Assuming the reaction:

$$^{235}_{92}U \rightarrow ^{148}_{57}La + ^{87}_{35}Br \tag{4.1}$$

one can calculate the mass defect, using the semiempirical Bethe–Weizsäcker equation (1,2,3). According to this formula, the physical atomic weight M of a nucleus, having Z protons and N neutrons is given as follows:

$$M(Z,N) = 1.01464\,A + 0.014\,A^{2/3} - 0.041905\,Z_A + \frac{0.041905}{Z_A}(Z - Z_A)^2 + \lambda\frac{0.036}{A^{3/4}} \tag{4.2}$$

where $A = Z + N$

$$Z_A = A/(1.980670 + 0.0149624\,A^{2/3})$$

$\lambda = +1, -1$ or 0 for respectively A even and Z odd, A even and Z even, A odd.

Applying equation (4.2), the masses of the isotopes occurring in the above hypothetical fission reaction are:

^{235}U : 235.11240

^{148}La : 147.98930

^{87}Br : 86.95722

Hence one can evaluate the mass defect of the fission reaction: 235.11240 $-(147.98930 + 86.95722) = 0.16588$ a.m.u., or $0.16588 \times 931 = 154$ MeV.

One could wonder, why under such exoergic conditions, spontaneous fission of the uranium nucleus does not occur. The inhibition is caused by the Coulomb barrier that both fission fragments have to overcome (see Chapter 3, section I). Using equation (3.30) and (3.1b) calculation of the barrier energy for the given example yields 197 MeV, when the surfaces of the La and the Br nuclei are in contact with each other. The barrier energy being about 43 MeV larger than the mass difference, spontaneous fission should not occur.

However, when a thermal neutron is absorbed, the binding energy of this particle increases the energy content of the uranium nucleus sufficiently to separate the two fission products:

$$^{235}U + ^{1}n \rightarrow ^{148}La + ^{86}Br \tag{4.3}$$

Application of equation (4.2) would yield a mass difference of 159 MeV, which is still insufficient to produce fission. The semiempirical mass formula neglects however the individual mass variation occurring at nuclei having "magic numbers" of nucleons, resulting in anomalously low masses. Thus, the energy released in the neutron induced fission (4.3) is in fact about 200 MeV, which permits the fragments to overcome the barrier energy.

Quantum mechanical treatment of the fission reaction predicts that for ^{235}U spontaneous fission occurs, but at such a low rate that the stability of the nucleus is practically not affected. Segré (4) reviewed theory and experiments about spontaneous fission, and noted that the mean life of the ^{235}U nucleus would be 3×10^{17} year, in the absence of α-emission. The reason for this spontaneous reaction is that the probability of fission fragments "tunneling" through the potential barrier is not zero. This is the same phenomenon as one encounters with charged particles, which can enter or leave a nucleus, although their kinetic energy is lower than the Coulomb barrier energy.

Using the liquid drop model of the nucleus, subject to the short-range nuclear forces, represented by surface tension, and to the long range Coulomb forces, an estimate of mass A and charge Z of a nucleus, which will be unstable against spontaneous fission can be found (5):

$$\frac{Z^2}{A} \geq 44 \tag{4.4}$$

where Z^2/A is the fissionability parameter.

This means that nuclei with a Z^2/A value clearly smaller than 44, will be stable against spontaneous fission, whereas nuclei having a Z^2/A value larger than 44 can hardly exist in nature. In Figure 4.1 the fissionability parameter is plotted against Z for several nuclei. Due to the tunneling effect, spontaneous fission practically occurs for all nuclei having a Z value above 90, but not as a major mode of the decay. In fact the partial half-lives for spontaneous fission of the even Z heavy nuclei are systematically decreasing as a function of increasing fissionability parameter, as can be seen from Figure 4.2 (6,7,8).

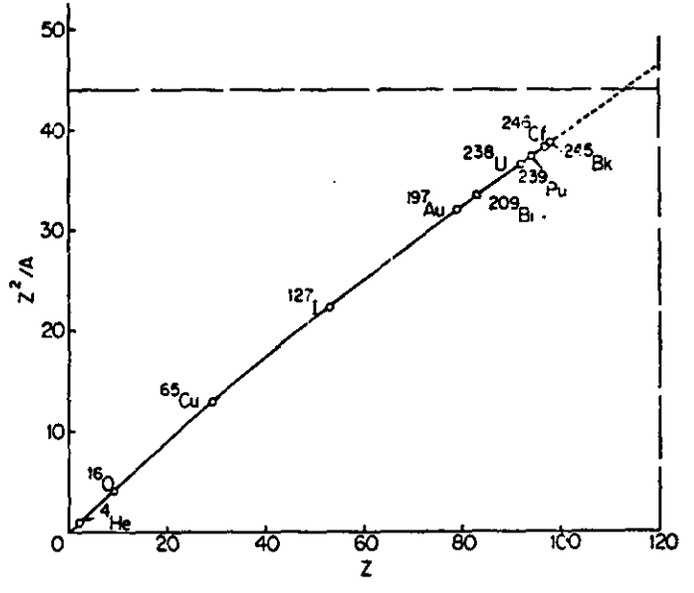

Fig. 4.1. The fissionability parameter Z^2/A, as a function of Z. (From *The Atomic Nucleus* by Evans, R. D. Copyright McGraw-Hill Inc. 1967. Used with permission of McGraw-Hill Book Company).

From this figure it also appears that even-even nuclei give rise more easily to spontaneous fission than even-odd nuclei. This is due to the fact that even-even nuclei are slightly lighter than even-odd ones, as can be calculated from the semiempirical mass formula (4.2). Therefore, fission by thermal neutron capture will be readily induced in even-odd nuclei, as the binding energy of the neutron in the reaction

$$\text{(even-odd) nucleus} + \text{n} \longrightarrow \text{(even-even) nucleus} \xrightarrow{\text{fission}}$$

is quite large, due to the relatively high mass difference between the compound nucleus and the absorbing nucleus. For this reason, $^{235}_{92}$U gives rise to fission by absorption of thermal neutrons, whereas $^{238}_{92}$U requires energetic (fast) neutrons for the same reaction. This is reflected in the thermal fission cross section for the uranium isotopes, as can be seen from Table 4.1.

The fission reaction with thermal neutrons is normally called asymmetric, as two fission products of unequal mass are obtained. However,

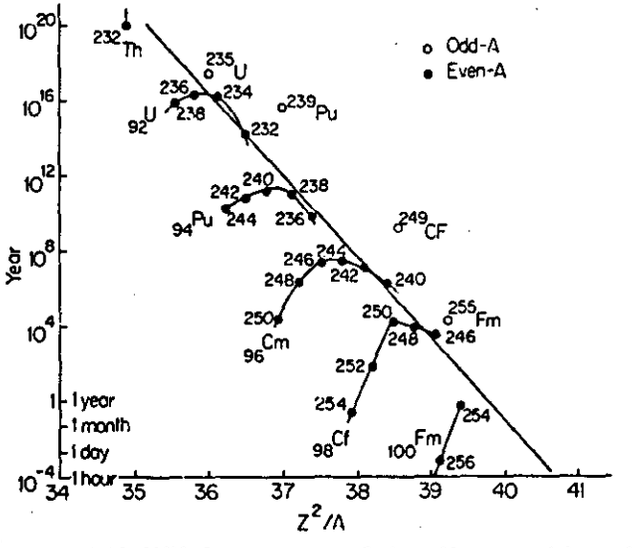

Fig. 4.2. Partial half-life for spontaneous fission of heavy nuclei as a function of Z^2/A. (From *The Atomic Nucleus* by Evans, R. D. Copyright McGraw-Hill Inc. 1967. Used with permission of McGraw-Hill Book Company).

TABLE 4.1
Thermal fission cross section for the uranium isotopes (11, 12)

Isotope	Type		σ_{fission} (barn)
	Z	N	
$^{233}_{92}$U	even-odd		527
$^{234}_{92}$U	even-even		20.65
$^{235}_{92}$U	even-odd		577
$^{236}_{92}$U	even-even		< 0.5 mb
$^{238}_{92}$U	even-odd		14

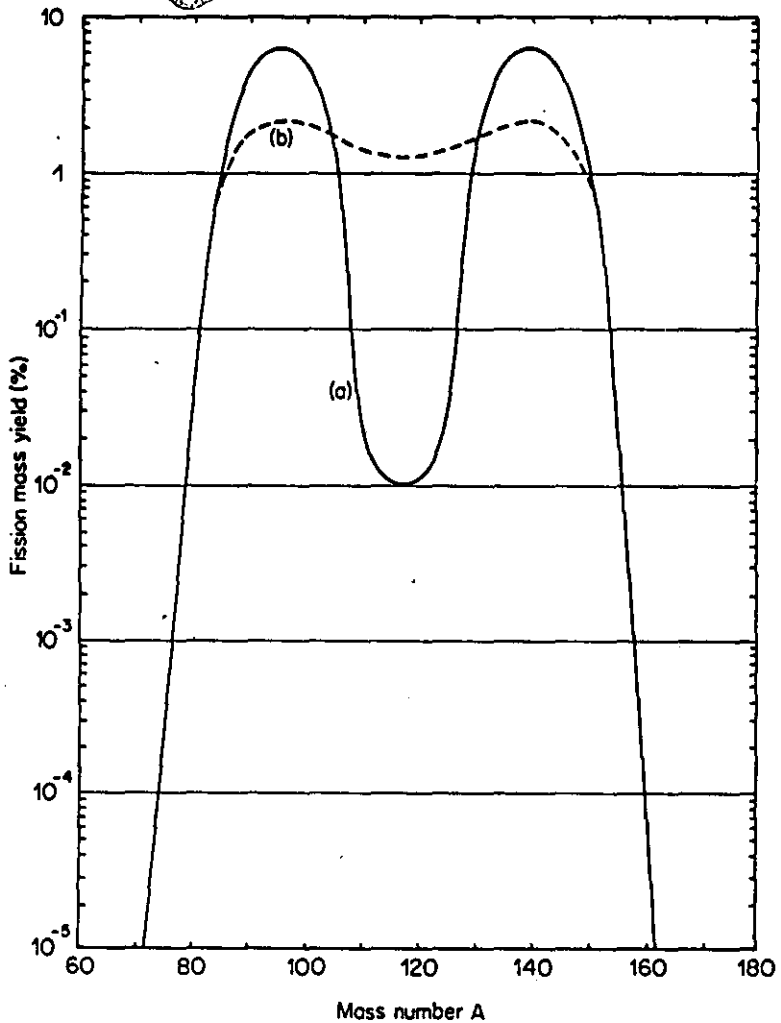

Fig. 4.3. Fission yield as a function of mass number A, for the thermal ^{235}U fission (a) and the 37 MeV alpha particle induced fission of ^{232}Th (b) (9) (10). (From *The Atomic Nucleus* by Evans, R. D. Copyright 1967 McGraw-Hill Inc. Used with permission of McGraw-Hill Book Company).

when highly energetic particles induce fission the reaction becomes more symmetrical. This is illustrated in Figure 4.3, where the fission yield curve is represented for two reactions, going over the same compound nucleus: (9,10)

$$^{235}_{92}U + {}^{1}_{0}n_{\text{thermal}} \longrightarrow [^{236}_{92}U] \xrightarrow{\text{fission}} \quad (4.5)$$

$$^{232}_{90}Th + {}^{4}_{2}\alpha_{(37\ \text{MeV})} \longrightarrow [^{236}_{92}U] \xrightarrow{\text{fission}} \quad (4.6)$$

As the heavy nuclei have a higher neutron excess than the light ones, the produced fission products tend towards proton–neutron equilibrium. Therefore already during the fission reaction several neutrons (prompt fission neutrons) are emitted, giving the typical equation:

$$^{235}_{92}U + {}^{1}_{0}n_{(\text{thermal})} \longrightarrow {}^{A_1}_{Z_1}X + {}^{A_2}_{Z_2}Y + \nu\, {}^{1}_{0}n \quad (4.7)$$

The number of neutrons ν is 2.5 for the ^{235}U fission and 2.9 for the ^{239}Pu fission. This prompt neutron emission is insufficient to establish the proton neutron equilibrium in the fission fragments. This equilibrium is eventually reached by several successive beta emissions of the created isotopes, of which a typical example:

$$^{137}_{52}Te \xrightarrow[1\ \text{m}]{\beta^-} {}^{137}_{53}I \xrightarrow[22.5\ \text{s}]{\beta^-} {}^{137}_{54}Xe \xrightarrow[3.4\ \text{m}]{\beta^-} {}^{137}_{55}Cs \xrightarrow[27\ \text{y}]{\beta^-} {}^{137}_{56}Ba\ \text{stable} \quad (4.8)$$

About 200 fission products of ^{235}U are known, which are practically all beta emitters, and show half-lives varying from unmeasurably short ones up to 10^6 year. The most abundant fission products, obtained by thermal fission of ^{235}U, have masses round the fission yield curve maxima (A = 95 and 140), and are given in Table 4.2. (53).

In some cases, the fission products emit a neutron immediately

TABLE 4.2
Most abundant fission products from the ^{235}U thermal fission (53)

Isotope	Half-life	Percentage fission yield
^{90}Sr	28.1 y	5.8
^{91}Y	58.8 d	5.4
^{95}Zr	65 d	6.4
^{99}Tc	2.12×10^5 y	6.0
^{106}Ru	39.6 d	3.0
^{137}Cs	30.0 y	6.15
^{140}Ba	12.8 d	6.3
^{144}Ce	284 d	6.0
^{147}Pm	2.62 y	2.7

after beta decay. This occurs when the excited state of the daughter nucleus has a larger potential energy than the binding energy of a neutron. This is often the case when the parent isotope has a "magic number" of protons. One can remark that without these so-called "delayed neutrons" the control of a nuclear reactor would never be possible, as will be discussed in the next section.

(B) Chain Reaction and Criticality

In all reactors, the average energy of the neutrons giving rise to fission is lower than the average energy with which they are produced. From Figure 4.4, where capture and fission cross sections of the isotopes ^{235}U and ^{238}U are represented (11,12), it appears that the fission probability for ^{235}U is highest with thermal neutrons (about 0.025 eV), whereas fission with ^{238}U only occurs with neutrons having an energy over 1 MeV. At neutron energies above 5 MeV, the fission cross section of ^{235}U and ^{238}U are of the same order of magnitude. The fission cross section for the two isotopes is always smaller than the one for ^{235}U with thermal neutrons. In order to use this reaction with optimum yield, the fuel can be enriched with ^{235}U, and the energetic fission neutrons (up to 10 MeV) can be slowed down by means of a moderator, so that thermal energy is reached before capture by ^{238}U.

In "fast" reactors, the average energy of the neutrons giving rise to fission is quite high, and the neutrons are slowed down by elastic collisions, mainly in the fuel elements and the core materials and to a smaller extent in the coolant (e.g. liquid sodium).

In "thermal" reactors, the fuel elements are surrounded by a moderator (light nuclei), which allows the neutrons to reach an average energy equal to the thermal motion energy of the atoms in the reactor core, before they can induce a new fission. The average distances traveled by a fission neutron before reaching thermal energy in some typical moderator materials is 5.7 cm in H_2O, 11.0 cm in D_2O, 9.9 cm in Be and 18.7 cm in graphite.

When the reactor medium is infinite, the reproduction factor k is defined as the ratio of the number of neutrons in one generation to the number of neutrons in the former generation. This is the number of fissions produced by one single fission in the previous cycle.

The reproduction factor is the product of the following four factors (12):

The number of neutrons released per fission (2.5 for thermal fission of ^{235}U);

The contribution of the fast fission. As the fuel contains large amounts of ^{238}U (only 0.71% ^{235}U is present in nonenriched fuel) a small percent of the fission neutrons may cause fast fission of ^{238}U, before leaving the fuel element;

The probability of resonance escape. As can be seen from Figure 4.4, the total cross section of ^{238}U has very high resonance peaks due partly to capture. There are always a few percent fission neutrons captured during the slowing down process, when they reach this particular energy region.

The thermal utilization: when the neutrons reach thermal energies, some are absorbed by the moderator, by the uranium, by impurities in both of them, and by fission products themselves.

When for an infinite fuel–moderator pattern $k_\infty = 1$, a steady-state equilibrium is reached, and a self sustaining chain reaction occurs. The excess k or the excess reactivity of a reactor is given by $k_\infty - 1$. When

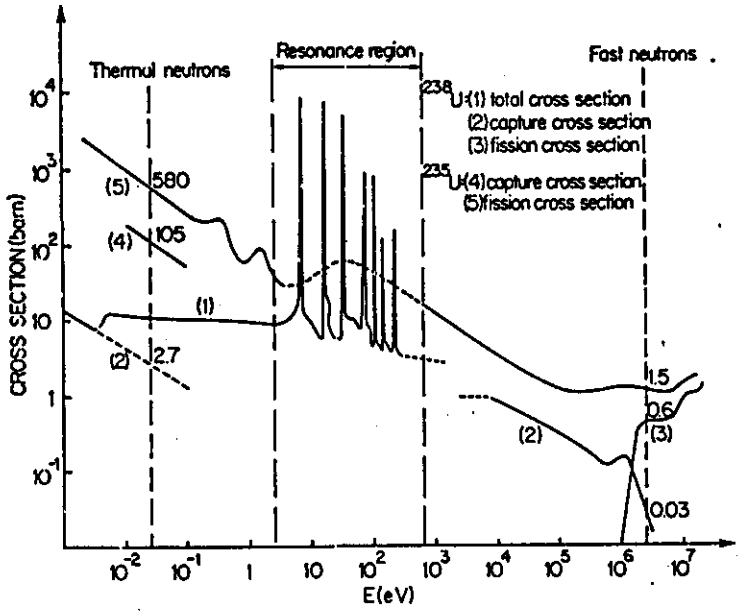

Fig. 4.4. Neutron capture and fission cross section of ^{235}U and ^{238}U (12).

$k_\infty > 1$, or the excess reactivity is positive, the number of neutrons, and thus the number of fissions increases with each generation, which can lead to an explosive reaction. When $k_\infty < 1$, or the excess reactivity negative, the number of neutrons decreases and the chain reaction dies out.

In practice however, the reactor arrangement is finite, which implies that some of the neutrons will escape from the core surface, decreasing in this way k_∞. The effective k (k_{eff}) of a reactor is given by k_∞ minus the leakage. When the neutron leakage equals the excess reactivity, a $k_{eff} = 1$ is obtained, and a steady-state equilibrium occurs again. The size of a bare finite reactor, for which this condition is fulfilled, is called the critical size. In the case of a graphite–uranium lattice, this results in a cube of about 6.5 m side. In order to reduce the dimensions one tries to minimize the neutron leakage by surrounding the pile with a reflector. Light nuclei such as graphite, hydrogen and beryllium are very suitable for scattering emerging neutrons back into the lattice. In this way the neutron leakage is reduced, and so is the critical size. However, the use of a reflector implies again the possibility of neutron capture either by the reflector atoms, or by impurities.

The steady-state equilibrium of the fission reaction is ensured in a reactor by inserting control rods composed of elements with high neutron capture cross sections, in such a way that k_{eff} equals unity.

In the starting procedure, however, a k_{eff} slightly over unity must be obtained in order to increase the operating level. The control rods are withdrawn, obtaining in this way a neutron excess, until the operating level is reached. A simple calculation shows that, if only prompt fission neutrons were present, an explosive chain reaction would occur. Assuming a k_{eff} of 1.001 (0.1% excess reactivity) the reactor period T_R, representing the time wherein the neutron flux increases by a factor e, is given by:

$$T_R = \frac{\tau}{k_{eff} - 1} \tag{4.9}$$

where τ is the average life-time of a thermal neutron. When only prompt fission neutrons are considered, $\tau \simeq 10^{-3}$ s, thus yielding a T_R of 1 s. This means that every second the neutron flux increases by a factor e or by a factor of about 20,000 every 10 seconds. To follow this rapid increase, very rapid control equipment and very quickly moving control rods are required, making the whole operation hazardous and

even impossible. However, the presence of about 1% of delayed neutrons, which are emitted several seconds after the fission has occurred, increases τ to about 0.1 s. Equation (4.9) yields then a reactor period T_R of 100 s, which allows ample time for adequate control, and ensures a start up without any danger.

(C) Classification of Nuclear Reactors

Since 1942, when the first graphite-uranium "pile" was constructed at the University of Chicago, reactors have been built using different geometrical forms, moderators, fuel elements and reflectors, and serving a wide variety of purposes. Classification of reactors can be made according to the following properties:

Type of fuel: The most simple fuel is natural uranium, having a composition of 6.10^{-3}% ^{234}U, 0.71% ^{235}U and 99.28% ^{238}U. To increase the amount of fissionable material, uranium enriched in ^{233}U, ^{235}U and ^{239}Pu is also used. Through the application of enriched uranium, which can range from a few percent up to 95% ^{235}U, a reduction in reactor size is obtained, because the number of fissions per gram fuel increases.

Type of moderator and coolant: As described in the previous section, the criterion, whether a reactor is fast or thermal, is completely dependent upon the fuel/moderator ratio. A moderator material must have the properties of large energy loss per neutron collision and small neutron capture cross section, in order to obtain an optimum neutron yield. These properties call for the use of very pure materials composed of light nuclei. Taking into account the costs of manufacturing a large quantity of very pure material, it is indicated to use water, heavy water, graphite or beryllium.

In addition to high purity, low neutron capture and low cost, the reactor coolant must show good properties of heat transfer. Air, water and liquid metals (Na, K) are used as coolants for nuclear reactors. In some constructions, as can be the case in swimming pool reactors, the moderator and the coolant are the same.

Neutron energy: As described in the previous section, most reactors use thermal neutrons to induce fission. Reactors using fast neutron energies can be applied in power plants to produce electrical energy and new fissionable materials.

Geometrical structure: A reactor is called heterogeneous when fuel and moderator are arranged separately from each other. In the case where fuel and moderator are homogeneousely mixed, a so-called homogeneous reactor is obtained. This type of reactor can be made by dissolving a uranium salt in water.

Purpose of the reactor: The purpose a reactor is built for, constitutes another mode of classification. A reactor can be built to produce a neutron excess for the formation of radioisotopes. An experimental or a research reactor serves as a study object for the construction of other reactors. Material testing reactors (MTR) have their main purpose in testing the resistance of materials, used in reactor construction, to radiation damage. Production of electrical energy can be ensured by a power reactor. When more fissionable material is produced than is burned up, the reactor is said to be a "breeder". Breeder reactors produce the fissionable isotopes ^{239}Pu and ^{233}U by the following reactions:

$$^{238}U + {}^{1}n \longrightarrow {}^{239}U \xrightarrow[23.5\,m]{\beta^-} {}^{239}Np \xrightarrow[2.3\,d]{\beta^-} {}^{239}Pu \qquad (4.10)$$

$$^{232}Th + {}^{1}n \longrightarrow {}^{233}Th \xrightarrow[23\,m]{\beta^-} {}^{233}Pa \xrightarrow[27\,d]{\beta^-} {}^{233}U \qquad (4.11)$$

From this classification it is clear that the distinction between reactors is arbitrary, but nevertheless it may be a useful guide. Summaries of the characteristics of many research reactors can be found in the literature (14,15,16,17,18).

(D) Reactor Neutron Fluxes

The fission neutrons slow down by elastic collisions with the moderator atoms until equilibrium is reached with the thermal motion of the moderator atoms (0.025 eV at 20°C). For a fission neutron of 2 MeV, the required number of collisions for this process is 18 with H, 25 with D, 114 with C, 150 with O and 2172 with U atoms. According to the degree of thermalization one generally distinguishes three different neutron spectra.

The unmoderated fission neutrons or the fast neutrons.

The energy of these neutrons is between 0 and 25 MeV for the ^{235}U fission. The fission flux $f(E)$ as a function of neutron energy E has been approximated by several semiempirical formulas:

$$f(E) = k_1 \exp(-E) \sinh \sqrt{(2E)} \qquad (4.12)$$

$$f(E) = k_2 \exp(-E/0.96) \sinh \sqrt{(2.29\,E)} \qquad (4.13)$$

$$f(E) = k_3 \exp(-E/1.29) \sqrt{E} \qquad (4.14)$$

where k_1, k_2 and k_3 are constants.

The Watt distribution (4.12) shows good agreement with the experimental data in the energy range from 0.075 to 15 MeV (19). Equation (4.13) was experimentally investigated between 0.18 and 12 MeV (20), whereas equation (4.14) gives a good approximation for the fission neutron spectrum below 9 MeV (21).

The slowing down or epithermal neutrons.

The neutrons, slowing down by elastic collision with the moderator atoms, show an energy distribution $\varphi_e(E)$ which varies as E^{-1}. This means that the epithermal neutron flux integrated over one logarithmic energy interval, can be represented by a constant φ_e, since:

$$\varphi_e(E) = \varphi_e E^{-1} \qquad (4.15)$$

It has to be noted that equation (4.15) only holds for a medium showing negligible neutron absorption.

The thermal neutrons.

Neutrons eventually reach thermal equilibrium with the moderator atoms and hence the thermal flux distribution $\varphi_{th}(E)$ as a function of energy is given by a Maxwell–Boltzmann distribution:

$$\varphi_{th}(E) = \frac{E}{E_m^2} \exp(-E/E_m) \qquad (4.16)$$

where $\varphi_{th}(E)$ is the neutron flux at energy E, normalized to unit integrated flux. $E_m = kT_m$ represents the neutron energy, corresponding to the most probable velocity v_0, and k is Boltzmann's constant.

It is obvious that the thermal neutron spectrum will be dependent on the reactor temperature. At 20°C the most probable velocity v_0, is 2200 m s^{-1} at a corresponding energy of 0.025 eV. The average velocity $\bar{v}$ is somewhat different from v_0, and is given by:

$$\bar{v} = (2/\sqrt{\pi}) v_0 \qquad (4.17)$$

In Figure 4.5 (22) the three different reactor neutron spectra are shown, normalized to unity integrated flux.

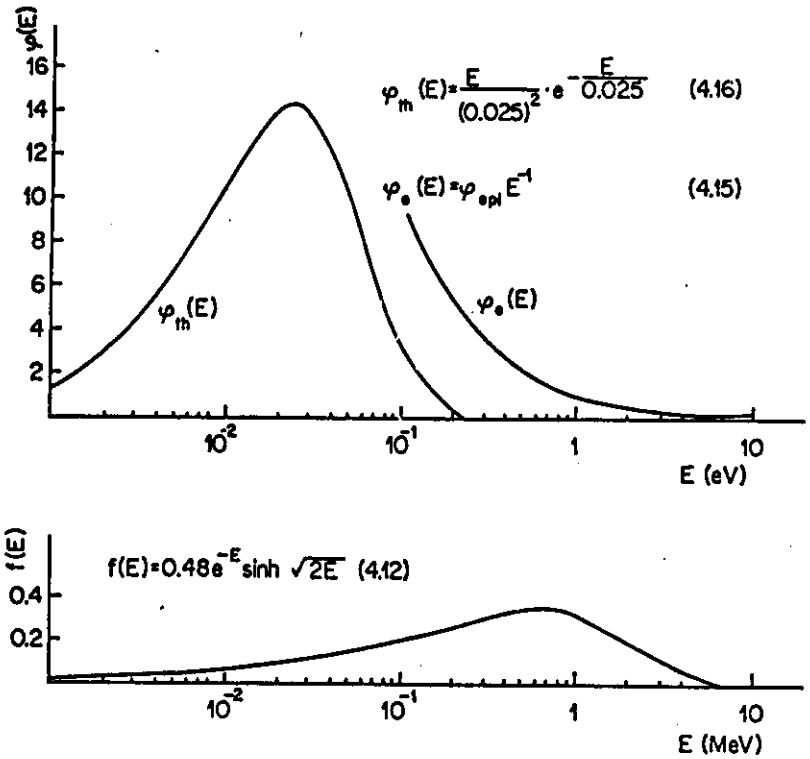

Fig. 4.5. Reactor neutron fluxes, normalised to unity integrated flux. (22)

In Chapter 3 it has been described how the (n, γ) reaction generally has a larger cross section than threshold reactions for the same isotope. As the former is induced by thermal neutrons, and the produced activity is proportional to the incident neutron flux (see Chapter 5), it is obvious that any reactor showing a reasonably high thermal neutron flux can be used for activation analysis. In order to avoid interferences, induced by threshold reactions (see Chapter 10), as pure a thermalized neutron spectrum as possible is desirable. Most isotope production reactors are equipped with a "thermal column", consisting of a pile of graphite blocks, adjacent to the reactor core. In this column, a more or less pure thermal neutron spectrum is obtained. A reasonably well thermalized flux can also be found in the reflector, although large gradients have to be taken into account. As an example, the fast to thermal flux ratios

together with the cadmium ratios are given for the BR–1 reactor (S.C.K. Mol–Belgium) in Table 3.8.

In some cases however, as described in Chapters 7 and 10, when activation analysis is performed by means of a threshold reaction, a pure fission spectrum is desired. This can be obtained by placing the sample in a hollow fuel element, where no moderator is present.

As described by Guinn *et al* (23,24) some types of reactors can be pulsed by quickly removing and reinserting the control rod. This procedure increases the excess reactivity, and neutron fluxes of the order of 10^{16} n cm^{-2} s^{-1} are obtained for a few milliseconds. This enhances the sensitivity for reactions, yielding short-lived isotopes as can be seen from Table 4.3.

TABLE 4.3
Calculated detection limits for some elements determined in a pulsed reactor, after Yule and Guinn (24)

Element	Reaction	Isotope produced	Half-life (s)	Measured photopeak (MeV)	Net photopeak activity (cpm/g) at end of irradiation t_e	Calculated detection limit (μg)*
O	n, p	^{16}N	7.35	6.1	2.1×10^7	48
F	n, γ	^{20}F	11	1.63	8.3×10^8	0.12
F	n, p	^{19}O	29	0.20	4.5×10^8	2.2
Na	n, p	^{23}Ne	38	0.44	1.4×10^8	7.0
Na	n, α	^{20}F	11	1.63	1.1×10^8	9.0
Mg	n, p	^{27}Na	60	0.38	4.8×10^8	210
S	n, p	^{32}P	12.4	2.1	1.0×10^8	1000
Sc	n, γ	^{46m}Sc	20	0.140	2.6×10^{12}	0.00035
Cr	n, p	^{51}V	225	1.44	5.0×10^8	20
As	n, p	^{75m}Ge	49	0.139	5.2×10^8	190
Se	n, γ n, n' n, 2n	^{77m}Se	17	0.160	5.0×10^{11}	0.002
Y	n, n'	^{89m}Y	16.1	0.915	9.5×10^8	0.10
Tb	n, 2n	^{159m}Tb	11	0.111	8.9×10^8	1100
W	n, γ n, n' n, 2n	^{183m}W	5.3	0.105	1.2×10^{10}	0.086
Pt	n, γ	^{199m}Pt	14	0.39	4.5×10^8	2.2
Au	n, n'	^{197m}Au	7.2	0.279	1.1×10^{11}	0.0089

* Detection limit defined by the authors as 10^3 photopeak cpm at $t = 0$ for $T_{1/2} < 1$ m, 100 photopeak cpm at $t = 0$ for $1 < T_{1/2} < 60$ m.

(E) Irradiation Facilities

As a consequence of the operation principle of a nuclear reactor, samples must be transferred in and out of the irradiation position by remote control. The materials used as sample packing, the irradiation can, the transfer container or "rabbit" and the rabbit tube will be exposed to radiation damage, and will become radioactive. The choice of adequate materials, depending on the kind of sample, the irradiation duration, the reactor neutron flux and the temperature, will be discussed in Chapter 7.

It is obvious that the rabbit tubes are cooled with the reactor coolant, and therefore in high flux reactors, one has to ensure good heat transfer between sample, packing, can, rabbit and rabbit tube. This can be achieved by filling the aluminium can with a solid pure aluminium block (99.95%), provided with holes for sample emplacement.

In order to obtain a reasonably homogeneous neutron flux over the whole rabbit, a careful choice of the rabbit tube emplacement with respect to the fuel element pattern is necessary. At the same time the fuel element arrangement must ensure a neutron flux as high as possible. In some reactors, samples are inserted into a ring around the core. Rotating this device one obtains the same neutron flux in each irradiation position. Plate 1 represents such a device, developed by Gulf General Atomic Inc. and called "Lazy Susan", with which Triga reactors can be equipped. The emplacement of the rabbit tube will evidently depend on the kind of neutrons desired. In-core irradiations will generally yield an appreciable fission neutron contribution, which in some cases can even be higher than the thermal flux. Activation in the thermal column and the reflector is mainly accomplished by thermal neutrons.

One can make a distinction between two major types of irradiation facilities: those that can be loaded and unloaded during reactor operation, and those that are only accessible when the reactor is closed down.

In the first case pneumatic transfer systems are the most widely used, in which the rabbits travel by either over or under pressure. Transfer times of the order of magnitude of one second can be achieved, which is of interest when dealing with very short half-lives. Typical rabbit tube emplacements around the reactor core are shown in Plate 2, for the isotope production reactor Thetis of Ghent University (Belgium.)

This reactor is mainly used for activation analysis purposes. To ensure adequate cooling, when irradiating in high flux reactors, a hydraulic transfer system can carry the rabbit in and out of the reactor. With swimming pool reactors, it is of course always possible to irradiate samples in the pool water. An aluminium cave containing the samples is attached to a nylon string. The cave is lowered into the pool, and the string fixed in such a way that the samples are adjacent to the core. Lead loading prevents the box from floating. It is clear that with a hydraulic system and in-pool irradiations, the cans must be watertight. A device which is particularly useful for isotope production and long irradiation times consists of a transport belt, looping through the reactor.

The off-operation facilities normally deal with irradiations in channels in the thermal column, in the reflector or between the fuel elements. These channels are hand loaded and access during operation becomes impossible. Hollow fuel elements belong to this type of facility and are used to obtain an undisturbed fission flux.

The problems of rabbit unloading after irradiation largely depend on the induced activity. For short irradiations and low flux reactors (10^{12} neutrons cm^{-2} s^{-1}) a normal glove box, provided with lead shielding of about 5 cm, is sufficient. With high flux reactors and long irradiations however, a hot cell becomes necessary to unpack the samples. Sometimes, between the irradiation position and the unloading station a shielded baby cave is built, where the short-lived activities are allowed to cool before treatment.

(F) Gamma Heating and Radiolysis

The most important source of heating in a nuclear reactor is caused by the absorption of gamma rays, to which the reactor neutrons give rise by several processes. One has to make a distinction between primary gammas already present in the reactor and the secondary gammas, produced in the sample during irradiation. The primary or core gamma flux is obtained by three effects, which give about equal contributions: the prompt fission gammas, released by fission of a uranium nucleus, the gammas produced by the decay of the fission products, and the prompt gammas emitted during thermal and resonance absorption of neutrons by the core materials, together with inelastic neutron scatter. It is obvious that the source strength of the core gamma source, expressed in MeV $cm^{-3}s^{-1}$, will be proportional

to the neutron flux. The heat development, normally given in watt per gram, in turn depends upon the gamma energy and the mass absorption coefficient of the materials in question, which depends on the density. The major part of the heat produced in the core is removed by the coolant, and thus the ambient temperature in the reactor will be dependent on the mechanical construction.

When a sample is introduced into a reactor, heating will be due to both primary and secondary gamma rays. Primary gamma rays will be absorbed in the sample and give rise to heat, which, when good heat transfer is provided, must be removed by the coolant. In this way the sample should reach thermal equilibrium with the reactor materials. On the other hand during activation, prompt gamma rays or particles are created in the sample by absorption and scatter of the neutrons. In addition radiation from the decay of the formed isotopes will also partly be absorbed in the sample. This can create a large temperature difference between the sample and the surrounding materials, to such an extent that low melting materials and alloys can liquify and that organic matter can carbonize.

The evaluation of the sample temperature is quite difficult as one deals with complex gamma spectra, perturbation of the neutron flux by the sample and gamma attenuation and build-up in the materials surrounding the sample. Temperatures can be experimentally determined by irradiating materials of known melting point or by using indicators, which change in color when a given temperature is reached. Even in this case, extreme caution in the conclusions drawn from the experiments has to be taken. The temperature attained in the material will not only depend on the ambient temperature but also on the neutron capture, which can become dominant when dealing with large absorption cross sections. As an example, tin, having a melting point of 232°C, will not melt during an irradiation of 1 week at 5×10^{12} neutrons $cm^{-2}\ s^{-1}$. Cadmium however, with higher melting point (320°C) will not withstand, because of the heating produced by the considerable neutron absorption. On the other hand, lead, with about the same melting point as cadmium (327°C) will not melt.

Schreiber and Allio (25) describe a method to estimate the gamma heating in materials placed in a reactor, provided the neutron fluxes are known. For this, they break up the gamma spectrum into two groups (0.8 and 2.5 MeV) and use empirical equations for the determination of the gamma attenuation and build up.

Taylor *et al* (26) evaluate the gamma heating of a sample irradiated in the core center of a 10 MW reactor, to be about 2.5 watt per g. An additional 3.5 watt is produced in the silica container and the aluminium can. As the sum of sample weights seldom exceeds 1 g, the total heat can be estimated to correspond with less than 6 watt. The authors calculated for the same reactor the nuclear heating for in-core irradiations of several cylindrical samples, with a volume of 1 cm^3 and a size of 20 × 8 mm diameter. The results are given in Table 4.4.

TABLE 4.4
Calculated nuclear heating in 1 cm^3 cylindrical samples (20 × 8 mm diameter), irradiated at core center in a 10 MW reactor (24)

Sample material	Heating (watt)
Al	7
Sb	17
Bi	25
Ca	4
Cr	18
Fe	20
Ni	24
Pt	60
Si	6
Na	2.5
Sn	16
W	70

When irradiating solutions, the sealed vials have to be leakproof for the pressure build up due to the ambient reactor temperature. In addition radiolysis also occurs giving rise to gases, which can build up considerable pressure. In pure water, H_2, O_2 and H_2O_2 are formed e.g. according to the reactions:

$$5\,H_2O \overset{h\nu}{\rightsquigarrow} H_2 + 2H_2O_2 + 3H^\cdot + {}^\cdot OH \qquad (4.18)$$

$$H^\cdot + O_2 \rightarrow HO_2^\cdot \qquad (4.19)$$

$$HO_2^\cdot + {}^\cdot OH \rightarrow O_2 + HO \qquad (4.20)$$

$$2HO_2^\cdot \rightarrow O_2 + H_2O_2 \qquad (4.21)$$

Similar reactions can give rise in HCl solutions to H_2 and Cl_2, whereas organic solvents produce even more complex reaction products

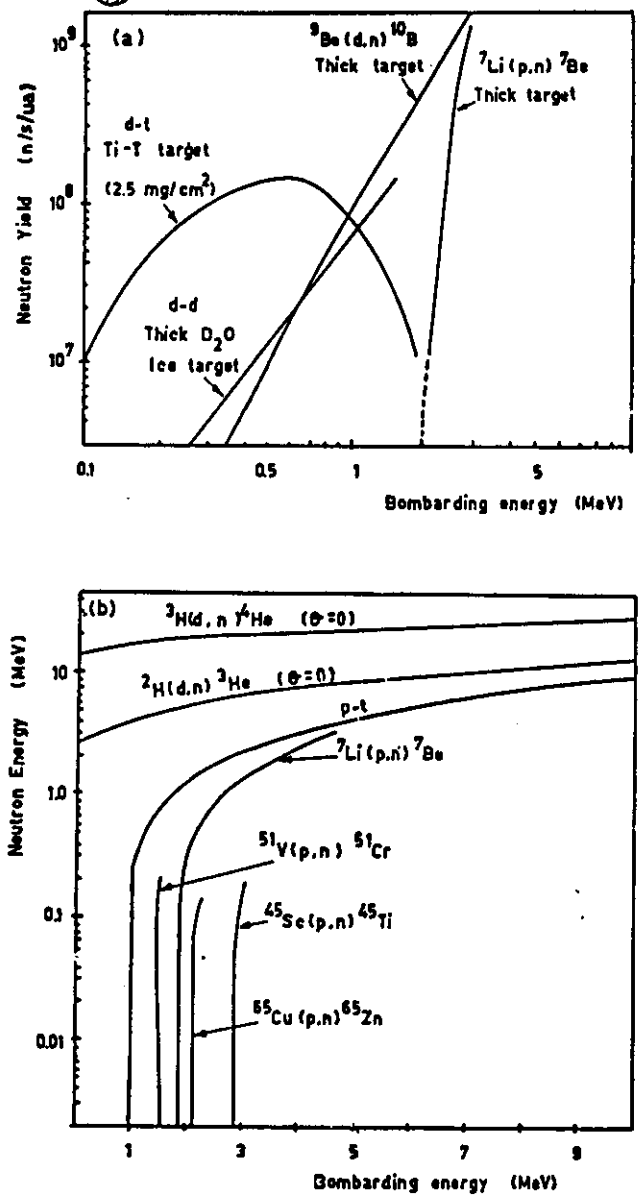

Fig. 4.6. Neutron yield (a) and neutron energy (b) as a function of bombarding energy for several deuteron and proton induced reactions (28).

(27). It is obvious that in many cases highly explosive gas mixtures under considerable pressure are obtained and that great care must be taken when opening the vials (see Chapter 7).

Because of this explosion hazard and the subsequent contamination possibilities, some authorities are reluctant to allow irradiation of solutions.

II. Neutrons from Accelerators

(A) Neutron Producing Reactions

In recent years, accelerator-produced neutrons have certainly taken their place among the irradiation sources. Theoretically a wide variety of neutron-producing reactions can be used, the choice of which will be highly dependent on the accelerator facility available (28). A further criterion in the choice of the reaction can be the cost per irradiation, which can be quite high with large accelerators. For convenience one can consider three groups of accelerating machines.

The first group contains the high energy charged particle accelerators, to which belong the Van der Graaf machine and the cyclotron. In a deuteron beam from these machines some of the exoergic (d, n) reactions commonly used are:

$$^{9}Be(d, n)^{10}B \quad (Q = 3.79\ MeV) \tag{4.22}$$

$$^{7}Li(d, n)^{8}Be \quad (Q = 15.0\ MeV) \tag{4.23}$$

$$^{2}H(d, n)^{3}He \quad (Q = 3.27\ MeV) \tag{4.24}$$

$$^{3}H(d, n)^{4}He \quad (Q = 17.6\ MeV) \tag{4.25}$$

The first reaction gives a very good neutron yield, as can be seen from Figure 4.6, although the obtained neutron spectrum is not monoenergetic and yields groups of energies between 1 and 6 MeV. The second reaction, giving rise to highly energetic neutrons, has however a neutron yield of about one third of that of the former one. The third reaction, sometimes called the d–d reaction, is normally performed on a D_2O ice target and yields almost monoenergetic neutrons. The last reaction, also called the d–t reaction, yields an isotropic source of nearly monoenergetic neutrons, and will be discussed in detail below.

Bombardment with energetic protons gives rise in most elements

to (p, n) reactions, whereby high neutron yields can be obtained. Some of the commonly used endoergic reactions are:

$$^{7}Li(p, n)^{7}Be \quad (E_T = 1.88\ MeV) \tag{4.26}$$

$$^{3}H(p, n)^{3}He \quad (E_T = 1.19\ MeV) \tag{4.27}$$

These reactions yield monoenergetic neutrons, the energy depending on the bombardment energy.

Figure 4.6, which has been adapted from Burrill and MacGregor (28) gives the neutron yield and the neutron energy as a function of the bombarding energy for several deuteron and proton induced reactions.

With alpha particles use can be made of the $^{9}Be(\alpha, n)^{12}C$ reaction ($Q = 5.6$ MeV) for the production of polyenergetic neutron groups between 1 and 6 MeV.

A second group of accelerators are the electron accelerating machines, producing Bremsstrahlung and subsequent (γ, n) reactions in the target material. This can be achieved with a betatron, a synchrotron or a linear accelerator (Linac), by stopping the beam in a high Z material. Thresholds of these (γ, n) reactions are located between 10 and 20 MeV. A few exceptions to this rule give an unusually low threshold, and therefore are sometimes used for neutron production. They are:

$$^{9}Be(\gamma, n)^{8}Be \quad (E_T = 1.66\ MeV) \tag{4.28}$$

$$^{2}H(\gamma, n)^{1}H \quad (E_T = 2.2\ MeV) \tag{4.29}$$

The neutron spectrum from these reactions is continuous and polyenergetic, due to the fact that the incident Bremsstrahlung shows a continuous energy distribution.

The combined (γ, n) and (γ, f) reaction in natural uranium has a high neutron yield, and produces a spectrum that is very analogous to the reactor fission spectrum.

All the above accelerators are mainly designed for physical experiments and are hardly accessible for activation analytical purposes.

A moderately expensive neutron generator can be obtained by using a 100 to 200 kV deuteron accelerator. A choice can be made between the $^{2}H(d, n)^{3}He$ or the $^{3}H(d, n)^{4}He$ reaction yielding respectively $\sim$ 2.6 MeV and $\sim$ 14 MeV neutrons. The latter is of more interest because at these low bombardment energies, monoenergetic neutrons of about 14 MeV are obtained with a yield 100 times higher than with the former. With the 14 MeV neutron source most threshold

reactions will occur, and can be used for activation analysis purposes, whereas the (n, γ) reaction gives an almost negligible contribution to the induced activity. The variation of neutron yield and energy, as a function of the angle θ with the incident deuteron beam is given in Table 4.5 (29).

TABLE 4.5
Neutron yield and neutron energy as a function of θ for the $^{3}H(d, n)^{4}He$ reaction. (29)

θ (degrees)	Neutron energy (MeV) for a deuteron energy E_D		Neutron yield relative to $\theta = 0°$
	E_D = 100 keV	E_D = 150 keV	
0	14.64	14.74	1.00
60	14.35	14.40	0.97
90	14.06	14.06	0.94
120	13.78	13.74	0.91
150	13.56	13.51	0.88
180	13.49	13.42	0.87

From Table 4.5 it appears that the source is nearly isotropic and monoenergetic. The excitation function of the reaction, measured with a thin target, stopping only 1 keV deuterons, reaches a maximum at 107 keV. In practice however, thick targets are used in order to stop the whole deuteron beam. In this case a maximal neutron production is situated at about 500 keV, as can be seen from Figure 4.6(a). From this figure it also appears that at 100 keV a reasonable neutron yield is still obtained.

The d–d reaction gives with this type of machine neutrons of about 2.6 MeV, which are not suitable to induce most threshold reactions, and on the other hand are too energetic to obtain an important (n, γ) contribution. The influence of the angle θ on the energy and the neutron yield is far more pronounced than with the foregoing reaction.

In general, one can state that in practically all cases fast neutrons are obtained, which can be used as such, or can be thermalized in order to induce (n, γ) reactions. Most attractive is the low energy deuteron accelerator, because of its low cost, its suitability for one person operation, and the possibility of using the fast neutron output

the irradiation site is achieved according to the "Texas convention" as described in Chapter 3, section VI, B, 4. It is obvious that steep gradients exist in the axial as well as in the transverse direction. The influence of this on neutron activation is discussed in Chapter 10, section II, B, 2.

When thermal neutrons are required, the target can be surrounded by a plastic, a paraffin or even a water moderator. A typical set up is obtained by placing the target in the middle of a cylindrical water tank with a diameter of about 1 m. Due to the large gradients however, the thermal neutron flux at the sample irradiation position is down by a factor of at least 10 in comparison to the fast neutron flux. Thus thermal fluxes are obtained, which are of the same order as the ones emitted by the isotopic neutron sources. The latter are preferable for the sake of stability.

(D) Irradiation Facilities

When using the neutron generator for activation analysis purposes, two problems have to be solved in order to obtain reproducible results. Firstly the inhomogeneity of the deuteron beam and the tritium target causes an inhomogeneous neutron flux, and secondly, the flux gradient through the sample gives rise to a gradient in the induced activity. As a partial solution to the first problem, rapid beam scanning devices have been proposed, causing a target irradiation in the same way as the electron beam scans a television picture tube. Generally, however, the irradiation facilities are built in such a way as to achieve a homogeneous exposure of the sample to the neutron flux, or so as to allow irradiation and measurement in strictly reproducible geometrical conditions. Because of the target depletion described in the previous section, it is obvious that neutron generators are most suited for short irradiations, hence for the formation of short lived isotopes. This implies a fast transport system, which can be achieved by pneumatic transfer. It is easily shown that in order to obtain fast travel times (of the order of one second) high flow rate at overpressure is the best solution.

As an absolute activation analysis method is very hard to perform, due to the many determining parameters, a relative analysis is generally adopted, where the element to be determined is measured versus a standard. In some procedures, standard and sample are irradiated

successively, and the neutron flux monitored during irradiation, in order to allow corrections for differences in neutron output. Flux monitoring can be achieved either by measuring the induced activity in a suitable element, which is irradiated simultaneously as well with the sample as with the standard, or by direct neutron counting during irradiation.

In the first case frequent use is made of the reaction $^{63}Cu(n, 2n)^{62}Cu$ ($T_{1/2}$ = 9.9 m), and the ^{62}Cu annihilation gamma rays are measured. With this method, care has to be taken that the degradation of neutron energy does not affect the activity ratio of the sample/monitor or the standard/monitor. This can occur when the cross section versus neutron energy curves of the monitor and the element to be determined have very different shapes and (or) thresholds.

Direct neutron counting can be achieved either with an organic plastic scintillator bead (38,39,40) or by means of a BF_3 counter (41,42,43,44) the latter surrounded by about 8 cm of paraffin in order to thermalize the neutrons before counting. This solution is especially valuable because of the almost complete insensitivity of the BF_3 counter for the important gamma flux associated with the neutron production. It has to be noticed that, when dealing with short lived isotopes, neutron flux monitoring has to take into account the decay of the isotope during irradiation (38,41). It is obvious that a burst of neutrons at the beginning of the irradiation will affect the measured sample activity to a smaller extent than a burst occurring at the end of the irradiation. Therefore, a ratemeter is sometimes branched in the neutron detection chain, having a time constant equal to the mean life ($1/\lambda = T_{1/2}/\ln 2$) of the isotope to be determined. In order to avoid these inconveniences, systems are constructed, wherein sample and standard are irradiated simultaneously.

To obtain homogeneous irradiations of the samples and standards, basically two systems have been described. In the first system the cylindrical sample is rotated around its longitudinal axis, either mechanically, or by means of an air jet, entering the rabbit tube tangentially (38,40). The set up can be performed with one tube only, when sample and standard are irradiated consecutively, or with two adjacent tubes, when both sample and standard are irradiated simultaneously. It has to be noticed that this system takes care of the axial flux gradient, but not of the transverse flux gradient. The second system consists of a dual sample biaxial rotator, ensuring that sample and

standard are irradiated simultaneously (45,46,47). The rotation is performed not only along the longitudinal axis, but sample and standard orbit round the deuteron beam axis. Although with this set up a somewhat smaller average flux is obtained in the irradiation position, due to greater sample–target distance, a very homogeneous irradiation is obtained. This can be of importance when high precision is desired, e.g. in the elementary analysis of chemical compounds. It has been described how a there and back corkscrew motion of the sample along an axis perpendicular to the beam achieves about the same reproducibility (48).

A system which allows irradiation and measurement of sample and standard under strictly constant geometrical conditions has been built by means of rabbit tubes of rectangular section (35). At the irradiation site, the tubes are placed one after another, the sample being nearest to the tritiated target. This assembly shows the advantage that no moving parts are present, thus increasing the reliability, and making it very suitable for use in industry. The relative neutron fluxes in the sample and standard position can be determined by means of two standards, and taken into account by the evaluation of the results. A schematic representation of the four systems is given in Figure 4.8.

In view of the large flux gradients, it is quite obvious that the irradiation facility has to be as close to the target assembly as possible. This can cause some inconvenience when the target is not at earth potential but at the negative accelerating voltage, as is sometimes the case in smaller "sealed" tubes. Indeed, in most rabbit systems metallic terminals are used, and with a negative accelerating voltage one has to enlarge the target–rabbit tube distance for the sake of insulation.

In all rabbit systems quite large samples are handled, in order to compensate for the rather low neutron flux and the small reaction cross sections. Therefore one has to take into account not only the flux gradients but also the neutron removal out of the beam, as is described in Chapter 10, section II, B, 4.

III. Isotopic Neutron Sources

Neutrons can be obtained by (α, n) or (γ, n) reactions on light nuclei, as described in section II of this chapter. When the bombarding particles or gamma rays are produced by the decay of a radioisotope, so called isotopic neutron sources are obtained. The alpha emitting

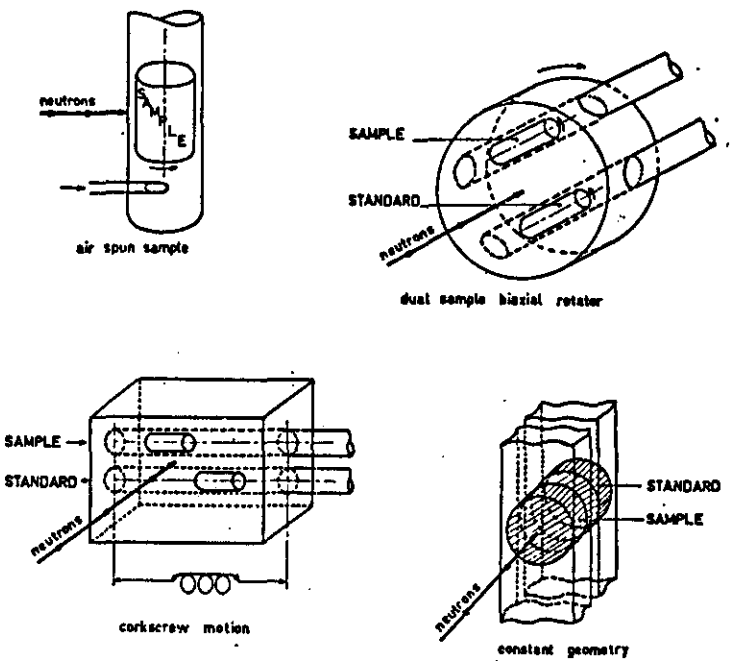

Fig. 4.8. Schematic representation of different irradiation facilities with a neutron generator.

isotopes that can be incorporated in this type of source are: ^{227}Ac, ^{241}Am, ^{210}Po, ^{226}Ra, ^{228}Th and ^{239}Pu. As gamma source ^{124}Sb is mainly used, although others can be applied, such as ^{24}Na, ^{88}Y and ^{140}La. The only requirement of the bombarding sources is that the emitted radiation has an energy above the reaction threshold.

As however the thresholds of these reactions are rather high, only light nuclei can be used as target material. The energy of the alpha particles, produced by the above mentioned isotopes, ranges between 4 and 6 MeV. Therefore beryllium is mainly used as a target element, because of its low (α, n) reaction threshold. With a lower neutron yield, boron, lithium and even fluorine can also be applied. Most of the alpha source isotopes emit several alpha rays of different energies and give rise to daughter isotopes, showing the same decay mode. Furthermore, the alpha particle energy is more or less degraded before reaction occurs, due to energy loss in the medium. On the other hand, the

product nucleus is not always obtained in the ground state. For these reasons it is quite obvious that the neutron spectrum will be far from monoenergetic, as can be seen from Figure 4.9, where the neutron spectra of a ^{226}Ra/Be, ^{241}Am/Be (49), and a ^{210}Po/B (50) source are represented.

As a consequence of the small penetrating power of an alpha particle, the neutron output will largely depend on the intimate mixing of the source and target material.

Practically all gamma induced reactions have a threshold energy larger than 8 MeV. Because of this, only two reactions can generally be applied:

$$^{9}\mathrm{Be}(\gamma, \mathrm{n})^{8}\mathrm{Be} \rightarrow 2^{4}\mathrm{He} \qquad (E_T = 1.67\ \mathrm{MeV}) \qquad (4.30)$$

$$^{2}\mathrm{H}(\gamma, \mathrm{n})^{1}\mathrm{H} \qquad (E_T = 2.23\ \mathrm{MeV}) \qquad (4.31)$$

Again beryllium is mainly used as a target material and so to a lesser extent is deuterium. As gamma rays are far more penetrating than alpha

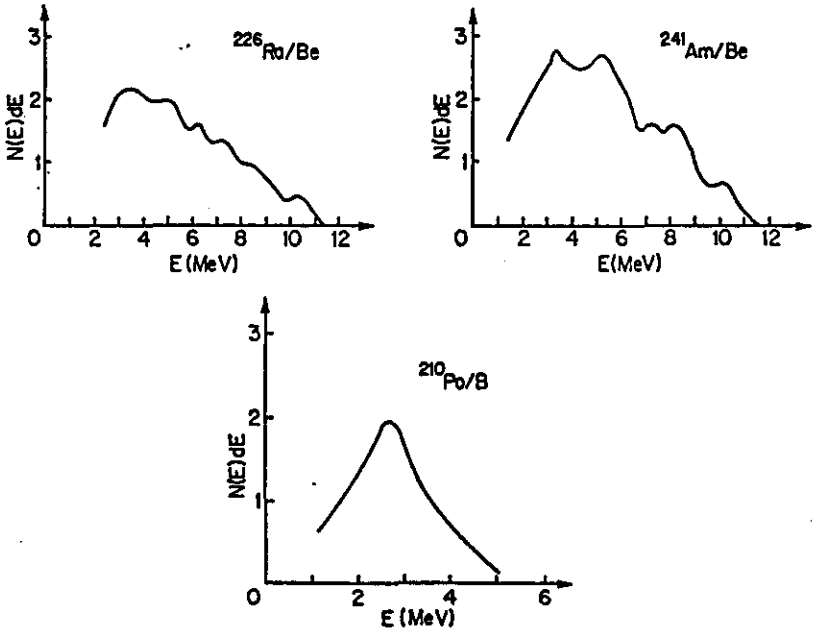

Fig. 4.9. Neutron spectra of the isotopic sources ^{226}Ra-Be (a), ^{241}Am-Be (b) and ^{210}Po-B (c).

rays, the gamma source and the target material can be kept separated, which facilitates the reloading of the source, and the adaption to the desired neutron output. An experimental set up has been described, in which a beryllium block loaded with ^{124}Sb sources was immersed in water. Thermal fluxes of about 6.10^{8} neutrons cm^{-2} s^{-1} were reported for a 9000 Ci ^{124}Sb loading (51). This useful irradiation facility handles up to six samples, each with a net effective irradiation volume of about 20 cm^{3}. Samples are transferred by means of a pneumatic rabbit system.

The fact that isotopic neutron sources can be made portable, and give rise to an extremely stable neutron flux, are certainly major advantages. On the other hand, the thermal neutron output is still low in comparison to a nuclear reactor, but can be of the same order

TABLE 4.7
Isotopic neutron sources

Source isotope	Half-life	Target	Neutrons s^{-1} Ci^{-1} emitted
^{227}Ac	22 y	Be	1.5×10^{7}
^{226}Ra	1620 y	Be	1.3×10^{7}
^{228}Th	1.9 y	Be	2×10^{7}
^{239}Pu	2.4×10^{4} y	Be	1.4×10^{7}
^{241}Am	458 y	Be	2.5×10^{6}
		B	5×10^{6}
		F	1.5×10^{5}
		Li	4×10^{6}
^{210}Po	138 d	Be	2.5×10^{6}
		B	5×10^{5}
		F	1.5×10^{5}
		Li	4×10^{6}
^{124}Sb	60 d	Be	1.9×10^{5} (1)
^{24}Na	15 h	Be	1.3×10^{5} (1)
		D_2O	2.7×10^{5} (1)
^{88}Y	104 d	Be	10^{5} (1)
		D_2O	3×10^{3} (1)
^{140}La	40 h	Be	3×10^{3} (1)
		D_2O	7.4×10^{3} (1)

(1) The neutron yield for (γ, n) reaction is reported for a 1 g target, placed at 1 cm from the gamma source.

of magnitude as the useful thermal flux obtained with a neutron generator. The quite significant gradient that occurs in the environment of the source imposes a very rigorous calibration of the fluxes in sample and standard irradiation positions. It is obvious that the flux gradient is largely influenced by the source-moderator pattern. As normally large samples are irradiated, one has to take into account the flux gradient in the sample and the self absorption by the sample (see Chapter 10).

A survey of the characteristics of some widely applied isotopic neutron sources is given in Table 4.7.

It has to be noticed that spontaneous fission of some artificially produced transuranium isotopes could be applied for the production of small neutron sources. A 10 μg quantity of ^{252}Cf ($T_{1/2} = 2.6$ y) emits about 3.10^7 neutrons s^{-1} by spontaneous fission. The mean energy of the neutrons is 1.5 MeV. The supply of this isotope is however very limited, but can perhaps in the future be more easily obtained.

References

1. Bethe, H., and Backer, R. F., *Rev. Mod. Phys.*, 8, 82 (1936).
2. Weizsäcker, C. F., *Z. Physik*, 96, 431 (1935).
3. Fermi, E., *Nuclear Physics*, p. 6, University of Chicago Press (1950).
4. Segré, E., *Phys. Rev.*, 86, 21 (1952).
5. Bohr, N., and Wheeler, J. A., *Phys. Rev.*, 56, 426 (1939).
6. Seaborg, G. T., *Phys. Rev.*, 85, 157L (1952).
7. Ghiorso, G., Higgins, G. H., Larsh, A. E., Seaborg, G. T., and Tompson, S. G., *Phys. Rev.*, 87, 163L (1952).
8. Hyde, E. K., *UCRL-9036 Rev.* (1962).
9. Plutonium Project Report, *Rev. Mod. Phys.*, 18, 513 (1946). *J. Am. Chem. Soc.*, 68, 2411 (1946).
10. Newton, A. S., *Phys. Rev.*, 75, 17 (1949).
11. Hughes, D. J., and Schwartz, R. B., *Neutron Cross Sections*, Brookhaven National Lab., Upton – New York (1958).
12. Hughes, D. J., Magurno, B. A., and Brussel, M. K., *Neutron Cross Sections, Supplement 1*, Brookhaven National Lab., Upton – New York (1960).
13. Hughes, D. J., *Pile Neutron Research*, Addison-Wesley Publ. Co. Inc., Cambridge 42, Mass. (1953).
14. I.A.E.A., Directory of Nuclear Reactors II, Research, Test and Experimental Reactors, 1–348, Vienna (1959).
15. I.A.E.A., Directory of Nuclear Reactors III, Research, Test and Experimental Reactors, 1–354, Vienna (1960).
16. Research and Testing Reactors the world around, *Nucleonics*, 20 (8), 116 (1962).

17. Iaben, H. S., *Nucleonics*, 10(3), 10 (1952).
18. Born, H. J., and Marcinowski, H. J., Kern Technik, Isotopen Technik und Chemie, 4 (12), 573 (1962).
19. Watt, B. F., *Phys. Rev.*, 87, 1037 (1952).
20. Cranberg, L., Frye, G., Nereson, N., and Rosen, L., *Phys. Rev.*, 103, 662 (1956).
21. Leachman, R. B., *Proc. Int. Conf. on Peaceful Uses of Atomic Energy*, I.A.E.A., Vienna, II, 193 (1955).
22. De Neve, R., *Proc. of the "Koninklijke Vlaamse Academie voor Wetenschappen, Letteren en Schone Kunsten van België"*, XXX, 2 (1968).
23. Lukens Jr., H. R., Yule, H. P., and Guinn, V. P., *Nucl. Instr. and Meth.*, 33, 273 (1965).
24. Yule, H. P., and Guinn, V. P., *Proc. Int. Conf. on Radiochem. Meth. of Anal.*, I.A.E.A., Vienna, II, 111 (1964).
25. Schreiber, R. E., and Allio, R. J., *Nucleonics*, 22(8), 120 (1964).
26. Taylor, C., West, R., and Whiting, M., *Proc. Int. Conf. on Production and Use of Short-lived Radioisotopes from Reactors*, I.A.E.A., Vienna, I, 67 (1963).
27. Spinks, J. W. T., and Woods, R. J., *An Introduction to Radiochemistry*, John Wiley and Sons, London (1964).
28. Burrill, E. A., and MacGregor, M. H., *Nucleonics*, 18(12), 64 (1960).
29. Prud'homme, J. T., Texas Nuclear Corporation Neutron Generators, 5820 – Austin, Texas (1962).
30. Moak, C. D., Reese Jr., H., and Good, W. M., *Nucleonics*, 9(3), 20 (1951).
31. Penning, F. M., and Moubius, J. H. A., *Physica*, 4, 1190 (1958).
32. Keller, R., *Helv. Phys. Acta*, 22, 78 (1949).
33. Carr, B. J., *Nucleonics*, 18(12), 75 (1960).
34. Gow, J. D., and Ruby, L., *Rev. Sci. Instr.*, 30(5), 315 (1959).
35. Hoste, J., De Soete, D., and Speecke, A., The Determination of Oxygen in Metals by 14 MeV Neutron Activation Analysis, *Euratom Rept. EUR 3565.e* Brussels (1967).
36. Accelerator Targets designed for the Production of Neutrons, *Euratom Rept. EUR 1815.d, f, e* Brussels (1964).
37. Accelerator Targets designed for the Production of Neutrons, *Euratom Rept. EUR 2641. d, f, e* Brussels (1966).
38. Anders, O. U., and Briden, D. W., *Anal. Chem.*, 31, 287 (1964).
39. Girardi, F., Pauli, J., and Sabbioni, E., *Euratom Rept., EUR 2290. f.* Brussels (1965).
40. Vogt, J. R., and Ehmann, W. D., *Radiochim. Acta*, 4, 24 (1965).
41. Fujii, I., Muto, H., Ogawa, K., and Tani, A., *Journ. At. Energ. Soc. Japan* 5, 455 (1963).
42. Stallwood, R. A., Mott, W. E., and Fanale, D. T., *Anal. Chem.*, 36, 6 (1963).
43. Fujii, I., Miyoshi, K., Muto, H., and Shimura, K., *Anal. Chim. Acta*, 34, 146 (1966).
44. Guinn, V. P., and Wagner, C. D., *Anal. Chem.*, 32, 317 (1960).
45. Mott, W. E., and Orange, J. M., *Anal. Chem.*, 37, 1338 (1965).
46. Wood, D. E., Jessen, P. L., and Jones, R. E., *Pittsburg Conf. Anal. Chem. and Appl. Spec.*, Feb. (1966).

47. Lundgren, F. A., and Nargolwalla, S. S., *Anal. Chem.*, 40(4), 672 (1968).
48. Burns, F. C., Priest, G. L., and Priest, H. F., *Soc. Appl. Spec.* Chicago, June (1966).
49. Tompson, M. N., and Taylor, J. M., *Nucl. Instr. and Meth.*, 37, 306 (1965).
50. Cochran, R. G., and Henry, K. M., *Rev. Sci. Instr.*, 26, 761 (1955).
51. Downs, W. E., *Tran. Am. Nucl. Soc.*, 10(1), 83 (1967).
52. Evans, R. D., *The Atomic Nucleus*, McGraw-Hill, New York (1967).
53. Zysin, Y. A., Lbov, A. A., and Sel'chenkov, L. I., *Fission Product Yields and their Mass Distribution*, Consultants Bureau, New York (1964).

CHAPTER 5

GROWTH AND DECAY OF RADIOACTIVITY DURING AND AFTER IRRADIATION

I. Laws of Radioactive Decay – Exponential Law

(A) Simple Case

Consider the case of a radioactive nucleus (1) decaying into a stable nucleus (2):

$$(1) \xrightarrow{\lambda} (2)\ (\text{stab.}) \tag{5.1}$$

This case can be compared to a monomolecular reaction. The reaction rate (number of disintegrations per second, D) is proportional to N the number of atoms of (1) present:

$$\frac{dN}{dt} = -\lambda N = D \tag{5.2}$$

λ is a constant, characteristic of the particular radioactive species, and is called the decay constant, having the dimension of a reciprocal time, t^{-1}. The integral of this simple differential equation is given by

$$N(t) = N^0 \exp(-\lambda t) \tag{5.3}$$

Here, $N(t)$ represents the number of atoms (1) at a time t, N^0 the number at $t = 0$. The radioactive decay is thus governed by an exponential law. Combining (5.2) and 5.3) one can write:

$$\boxed{D(t) = D^0 \exp(-\lambda t)} \tag{5.4}$$

Measuring a fraction z of the real number of disintegrations (i.e. the experimentally observed activity A) one obtains:

$$A = zD = z\lambda N$$

and

$$A(t) = A^0 \exp(-\lambda t) \tag{5.5}$$

The coefficient z is called the detection coefficient and will depend on the nature of the detection instrument, the efficiency for the recording