فيزياء نووية

ثالثة تربية انجليزي (لائحة قديمة) الترم الثاني 2024/2023

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Chapter 1

ARTIFICIAL NUCLEAR DISINTEGRATION

11-1 Transmutation by alpha-particles: alpha-proton reactions. The fact that certain atoms undergo spontaneous disintegration led to speculation about the possibility of causing the disintegration of the ordinary inactive nuclides. It seemed possible that if atoms were bombarded with energetic particles, one of the latter might penetrate into a nucleus and cause a disruption. The radiations from the natural radionuclides could be used as projectiles, and α -particles seemed most likely to be effective because of their relatively great energy and momentum. Since most of the bombarding α -particles would probably be scattered, it was apparent that the probability of causing a nuclear disintegration would be small. The extent of the scattering could, however, be reduced and the probability of disintegration increased by using some of the lighter atoms as targets, thereby reducing the magnitude of the repulsive Coulomb forces between the target nuclei and the α -particles.

The first disintegration based on these ideas was made by Rutherford (1919),⁽¹⁾ who showed that the nuclei of nitrogen atoms emit swift protons when bombarded with α -particles from radium C. The apparatus that Rutherford used was simple but sensitive, and is shown schematically in Fig. 11-1. In one end E of a box B was cut an opening which was covered by a silver foil F. A zinc sulfide screen was placed at S, just outside the opening, and scintillations on the screen were observed by means of a microscope M. The source of the α -particles was radium C placed on a small disc D, whose distance from S could be varied. The silver foil F was thick enough to absorb the α -particles from the source. Different gases could be introduced into the box and removed through the side tubes T. When the box was filled with oxygen or carbon dioxide

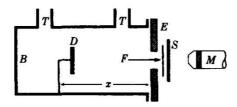


FIG. 11-1. Diagram of Rutherford's apparatus for the disintegration of nitrogen nuclei by α -particles. [Reprinted by permission from Rutherford, Chadwick, and Ellis, *Radiations from Radioactive Substances*, Cambridge University Press (Macmillan Co.), 1930.]

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at atmospheric pressure, no scintillations were seen on the screen with the source 7 cm or more away. This thickness x of gas was great enough to absorb all of the α -particles from radium C even without the silver foil. When the gas in the box was nitrogen, however, scintillations were observed on the screen when the source of the α -particles was as much as 40 cm away. Since it was known that α -particles from radium C could not penetrate 40 cm of air, Rutherford concluded that the scintillations were caused by particles ejected from the nitrogen nucleus by the impact of an α -particle. Measurement of the magnetic deflection of the particles suggested that they were protons, and this surmise was confirmed by more precise work.

Rutherford ruled out, by means of careful experiments, the possibility that the protons came from hydrogen present in the nitrogen as an impurity, and concluded that artificial disintegration of nitrogen atoms had taken place. The disintegration was caused by the α -particles from the radium C, and one result was the emission of a highly energetic proton by the nitrogen nucleus. The experimental results also showed that the probability of disintegration was very small; one proton was produced for about one million α -particles passing through the gas. Rutherford and Chadwick⁽²⁾ extended the work on nitrogen to other elements and found evidence of the disintegration of all of the light elements, from boron to potassium, with the exception of carbon and oxygen. They also found that in some cases the energy of the ejected protons was greater than that of the bombarding α -particles. This result provided additional evidence that the protons were emitted as the result of a disintegration process, the extra energy being acquired in the accompanying nuclear rearrangement.

Two hypotheses were suggested as to the nature of the nuclear process leading to the emission of the proton. They were: (a) The nucleus of the bombarded atom simply loses a proton as the result of a collision with a swift α -particle. (b) The α -particle is captured by the nucleus of the atom it hits, and the new, or compound, nucleus emits a proton. These two hypotheses for the disintegration process could be subjected to experimental test because in case (b) the α -particle should disappear. while in case (a) it should still exist after the collision. The choice between the two possibilities was settled in 1925 when Blackett⁽³⁾ studied the tracks produced by α -particles passing through nitrogen in a cloud chamber. He showed, as can be seen in Fig. 11-2, that as a result of a disintegration, the only tracks which could be seen were those of the incident α -particle, a proton, and a recoil nucleus. The absence of a track corresponding to an α -particle after the collision proved that the α -particle disappeared completely. If the disintegration process had been the result simply of a disruption leading to the emission of a proton from the nitrogen nucleus, there should have been four tracks rather than the three actually

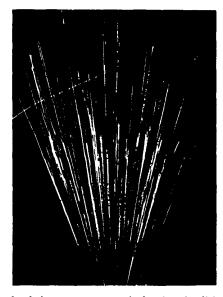


FIG. 11-2. A cloud chamber photograph showing the disintegration of a N¹⁴ nucleus by an α -particle with the formation of O¹⁷ and a proton. The long track of the proton and the short track of the recoiling oxygen nucleus can be seen. (Blackett.⁽³⁾)

seen. It was concluded, therefore, that the α -particle entered the nucleus of the nitrogen atom with the formation of an unstable system which immediately expelled a proton.

The disintegration of a nitrogen atom by an α -particle may be represented by an equation analogous to those used for chemical reactions,

$$_{7}N^{14} + _{2}He^{4} \rightarrow [_{9}F^{18}] \rightarrow _{8}O^{17} + _{1}H^{1}$$
.

In this equation, the symbols on the left stand for the reacting nuclides. The symbol in brackets stands for the unstable nucleus formed as the result of the capture of the α -particle by the nitrogen nucleus; this kind of nucleus is often called a *compound nucleus*. The emitted proton and the final nucleus, the products of the reaction, are on the right side of the equation. The charge and mass numbers must be the same on the two sides of the nucleus of an isotope of oxygen. In other words, an atom of nitrogen has been transformed, or transmuted, into an atom of oxygen. This transmutation may also be represented by the abbreviated notation N¹⁴(α ,p)O¹⁷. The transmutation of other nucleus by

 α -particles from radioactive substances may be represented by equations similar to that for nitrogen.

$${}_{5}B^{10} + {}_{2}He^{4} \rightarrow [{}_{7}N^{14}] \rightarrow {}_{6}C^{13} + {}_{1}H^{1}$$

$${}_{11}Na^{23} + {}_{2}He^{4} \rightarrow [{}_{13}Al^{27}] \rightarrow {}_{12}Mg^{26} + {}_{1}H^{1}$$

$${}_{13}Al^{27} + {}_{2}He^{4} \rightarrow [{}_{15}P^{31}] \rightarrow {}_{14}Si^{30} + {}_{1}H^{1}$$

$${}_{16}S^{32} + {}_{2}He^{4} \rightarrow [{}_{18}A^{36}] \rightarrow {}_{17}Cl^{35} + {}_{1}H^{1}$$

$${}_{19}K^{39} + {}_{2}He^{4} \rightarrow [{}_{21}Se^{43}] \rightarrow {}_{20}Ca^{42} + {}_{1}H^{1}$$

In each case, the charge of the nucleus is increased by one unit and the mass is increased by three units. The alpha-proton reaction may, therefore, be written in the form

$$_{Z}X^{A} + {}_{2}\text{He}^{4} \rightarrow [_{Z+2}Cn^{A+4}] \rightarrow {}_{Z+1}Y^{A+3} + {}_{1}\text{H}^{1},$$
 (11-1)

where X, Y, and Cn represent the target, product, and compound nuclei, respectively.

It is often convenient to refer to a nuclear reaction in terms of the incident and emitted particles, apart from the nuclei involved. Thus, the reactions mentioned so far are examples of (α, p) reactions.

11-2 The balance of mass and energy in nuclear reactions. A nuclear reaction such as that represented by Eq. (11-1) can be analyzed quantitatively in terms of the masses and energies of the nuclei and particles involved. The analysis of nuclear reactions is one of the main sources of information about nuclear properties and will therefore be discussed in some detail before more reactions are considered. The analysis is similar to that used for chemical reactions except that the relativistic relation between mass and energy must be taken into account. Consider a nuclear reaction represented by the equation

$$x + X \to Y + y, \tag{11-2}$$

where X is the target nucleus, x the bombarding particle, Y the product nucleus, and y the product particle. In the only type of reaction considered so far, x is an α -particle, and y is a proton. In other reactions, to be discussed in later sections of this chapter, other bombarding and product particles will be met. It will be assumed that the target nucleus X is initially at rest so that it has no kinetic energy. Since the total energy of a particle or atom is the sum of the rest energy and the kinetic energy, the statement that the total energy is conserved in the nuclear reaction means that

$$(E_x + m_x c^2) + M_X c^2 = (E_Y + M_Y c^2) + (E_y + m_y c^2).$$
(11-3)

In Eq. (11-3), m_x , M_X , m_y , and M_Y represent the masses of the incident particle, target nucleus, product particle, and product nucleus, respectively; the E's represent kinetic energies. We now introduce the quantity Q, which represents the difference between the kinetic energy of the products of the reaction and that of the incident particle.

$$Q = E_Y + E_y - E_z.$$
(11-4)

The quantity on the right side of Eq. (11-4) can be expressed in terms of the masses because of the relationship (11-3),

$$E_Y + E_y - E_x = (M_X + m_x - M_Y - m_y)c^2.$$
 (11-5)

Hence, from Eqs. (11-4) and (11-5),

$$Q = E_Y + E_y - E_z = (M_X + m_z - M_Y - m_y)c^2. \quad (11-6)$$

The quantity Q is called the *energy balance* of the reaction or, more commonly, the *Q-value*; it can be determined either from the energy difference or from the mass difference in Eq. (11-6).

If the value of Q is positive, the kinetic energy of the products is greater than that of the reactants; the reaction is then said to be *exothermic* or *exoergic*. The total mass of the reactants is greater than that of the products in this case. If the value of Q is negative, the reaction is *endothermic* or *endoergic*. It is apparent from Eq. (11-6) that the analysis of nuclear reactions involves information about nuclear masses and particle energies. Nuclear reactions can, therefore, be used to obtain information about the masses of nuclei, about particle energies, or about Q-values, depending on what information is available, and which quantities can be measured.

The term E_Y in Eq. (11-6) represents the recoil (kinetic) energy of the product nucleus. It is usually small and hard to measure, but it can

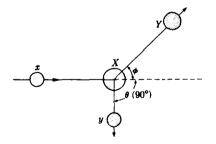


FIG. 11-3. Conservation of momentum in nuclear reactions. The outcoming particles are viewed at an angle of 90° with the direction of the incident particles.

be eliminated by taking into account the conservation of momentum. Consider, for simplicity, the special case in which the outcoming particles are observed at an angle of 90° with the direction of the beam of projectiles, taken as the *x*-axis, as in Fig. 11-3. Before the collision, the momentum vector is directed along the *x*-axis. Since the resultant momentum after the collision must also be directed along the *x*-axis, it follows that

$$m_x v_x = M_Y V_Y \cos \phi, \qquad m_y v_y = M_Y V_Y \sin \phi, \qquad (11-7)$$

where v_x , v_y , and V_Y are the velocities of the incident particle, the ejected particle, and the recoil nucleus, respectively, and ϕ is the angle between the *x*-axis and the direction of recoil of the product nucleus. When the last two equations are squared and added, the result is

$$(m_x v_x)^2 + (m_y v_y)^2 = (M_Y V_Y)^2$$

Setting $E_x = \frac{1}{2}m_x v_x^2$, $E_y = \frac{1}{2}m_y v_y^2$, $E_Y = \frac{1}{2}M_Y V_Y^2$, and solving for E_Y , we get

$$E_{Y} = \frac{m_{x}}{M_{Y}} E_{x} + \frac{m_{y}}{M_{Y}} E_{y}.$$
 (11-8)

Insertion of this expression for E_Y into Eq. (11-6) gives

$$Q = E_{y}\left(1 + \frac{m_{y}}{M_{y}}\right) - E_{x}\left(1 - \frac{m_{x}}{M_{y}}\right). \quad (11-9)$$

The value of Q can be determined, therefore, if the energies of the incident and ejected particles are measured and if the mass number of the product nucleus is known.

In the general case of a reaction in which the outcoming particle is observed at an angle θ with the direction of the projectile beam, it can be shown that (see Problem 19 at the end of the chapter),

$$Q = E_{y}\left(1 + \frac{m_{y}}{M_{Y}}\right) - E_{z}\left(1 - \frac{m_{z}}{M_{Y}}\right) - \frac{2}{M_{Y}}\left(E_{z}E_{y}m_{z}m_{y}\right)^{1/2}\cos\theta.$$
(11-10)

The importance of the last term decreases as the mass of the target nucleus increases. If the masses are not known accurately, a good approximation to the value of Q can be obtained by using the mass numbers instead of the actual masses in Eqs. (11-9) and (11-10). Equation (11-10) reduces, of course, to Eq. (11-9) when $\theta = 90^{\circ}$.

In an endoergic reaction, the energy -Q is needed to excite the compound nucleus sufficiently so that it will break up. This energy must be supplied in the form of kinetic energy of the incoming particle. But not all of that kinetic energy is available for excitation because some is used to impart momentum to the compound nucleus; this momentum is then distributed among the products of the reaction. Consequently, for -Q to be available for excitation of the compound nucleus, we must supply some energy in addition to -Q. The amount of energy needed for an endoergic reaction is called the *threshold energy* and can be calculated easily.

If we let M_C and V_C denote the mass and velocity of the compound nucleus, conservation of momentum requires that

$$m_z v_z = M_C V_C$$
, or $V_C = \frac{m_z}{M_C} v_z$.

The part of the kinetic energy of the incident particle needed for excitation of the compound nucleus is

$$-Q = \frac{1}{2} m_x v_x^2 - \frac{1}{2} M_C V_C^2 = \frac{1}{2} m_x v_x^2 \left(1 - \frac{m_x}{M_C}\right)$$

But, $M_C = M_X + m_z$, and

$$(-Q) = \frac{1}{2} m_x v_x^2 \left(\frac{M_X}{M_X + m_x} \right).$$

The threshold energy is then

$$E_{\rm th} = \frac{1}{2} m_x v_x^2 = (-Q) \left(1 + \frac{m_x}{M_x} \right); \qquad (11-11)$$

it can be determined experimentally and the result used to find the value of Q from Eq. (11-11). For a reaction induced by γ -rays, $m_x = 0$, and the threshold energy is just -Q.

The masses which appear in the equations of this section are nuclear masses. In actual calculations, however, they may be replaced by the masses of the neutral atoms. The electrons which must be added to the nuclei to form the neutral atoms cancel in the equation for a nuclear reaction because the number of electrons is the same on the two sides of the equation. As an example of the use of Eq. (11-6) consider the N¹⁴(α ,p)O¹⁷ reaction. The following values of the atomic masses are obtained from Table 11-1, Section 11-9.

$$M_X = M(N^{14}) = 14.007518$$
 amu
 $m_x = M(He^4) = 4.003873$ amu
 $M_Y = M(O^{17}) = 17.004529$ amu
 $m_x = m(H^1) = 1.008144$ amu

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The value of Q as calculated from the masses is Q = -0.001282 amu = -1.19 Mev and the reaction is endoergic. A Q-value of -1.16 ± 0.04 Mev has been obtained experimentally from energy measurements and agrees very well with the value calculated from the masses; according to Eq. (11-11), the threshold energy is 1.49 Mev.

The equation for a nuclear reaction sometimes includes Q; for example,

$$X + x \to [Cn] \to Y + y + Q.$$

Although this is not necessary, it is a convenient way of showing the energy balance.

11-3 The neutron: alpha-neutron reactions. The discovery of the neutron has been discussed briefly in Section 8-3; it will now be treated in somewhat greater detail. The capture of an α -particle by a nucleus does not always result in the emission of a proton by the compound nucleus. When beryllium was bombarded by α -particles, one of the products of the reaction seemed to be a very penetrating kind of radiation, and no protons were observed. It was assumed that the radiation consisted of γ -ray photons produced by the reaction

$$_{4}\text{Be}^{9} + _{2}\text{He}^{4} \rightarrow [_{6}\text{C}^{13}] \rightarrow _{6}\text{C}^{13} + \gamma.$$
 (11-12)

This assumption led, however, to difficulties. It was shown from absorption measurements that the energy of the "photons" should be about 7 Mev, but these radiations were able to knock protons out of hydrogenous materials and to impart to those protons energies of about 5 Mev. On the assumption that the protons were liberated by elastic collisions with γ -ray photons, calculations showed that each photon must have had an energy of about 50 Mev, a value much greater than that deduced from the absorption measurements. There was, therefore, a serious contradiction between the values of the photon energy given by the two methods. The energy that can be attained by the assumed γ -radiation can also be computed from known masses and energies. It is the sum of the *Q*-value for the reaction (11-12) and the kinetic energy of the incident α -particle minus the recoil energy of the product nucleus. The *Q*-value is given by the formula

$$Q = (M_X + m_x - M_Y)c^2,$$

since m_y , the mass of the photon, is zero. Inserting the values $M_X = 9.01504$, $m_x = 4.00387$, and $M_Y = 13.00748$, we get Q = 10.5 Mev. If the kinetic energy of the α -particle is assumed to be 5 Mev, the total energy available for the reaction is 15.5 Mev, part of which is retained as recoil energy of the product nucleus. The greatest possible energy of

the assumed γ -radiation is, therefore, less than 15.5 Mev, which is again much smaller than 50 Mev.

The problem of the identity of the products of the bombardment of beryllium was solved by Chadwick,⁽⁴⁾ as was mentioned in Chapter 8. In a series of experiments on the recoil of nuclei which were struck by the rays coming from the bombarded beryllium, he showed that if these rays were assumed to be γ -rays then the results of the experiments led to values for their energy which depended on the nature of the recoil nucleus. For example, protons ejected from paraffin had energies which led to a photon energy of 55 Mey, while recoiling nitrogen nuclei had energies which led to a photon energy of about 90 Mey. If the energy of the recoil nuclei was assumed to be produced by elastic collisions with photons, it turned out that the energy that had to be attributed to the photon increased with the mass of the recoil atom—a requirement that is contrary to the principles of conservation of energy and momentum in elastic collisions. Chadwick showed that all of the difficulties disappeared when the hypothesis was made that the radiation coming from beryllium bombarded with α -particles consists of particles (neutrons) with a mass very nearly equal to that of the proton, but having no charge. The nuclear reaction which produces these neutrons is

$$_{4}\text{Be}^{9} + _{2}\text{He}^{4} \rightarrow [_{6}\text{C}^{13}] \rightarrow _{6}\text{C}^{12} + _{0}\text{n}^{1},$$
 (11-13)

where $_{0}n^{1}$ is the symbol for the neutron. The penetrating nature of the neutrons follows from the absence of a charge, and the energies of the recoil atoms in Chadwick's experiments could be completely accounted for on the basis of elastic collisions with an energetic particle of unit mass.

Chadwick also proved that the mass of the neutron is approximately equal to that of the proton, by means of the experiments on the collisions of neutrons with protons and nitrogen nuclei. Since the neutron is not a charged particle, its mass could not be determined from deflection measurements in electric and magnetic fields, and indirect methods had to be used. Chadwick's method was based on the fact that in a collision between a moving and a stationary particle, the velocity imparted to the latter is greatest in a head-on collision, one in which the struck particle moves in exactly the same direction as that in which the incident particle approached it. An expression for the maximum velocity can be derived from the equations of conservation of kinetic energy and momentum in a head-on collision. Suppose that a particle with mass m_1 and velocity vcollides with a stationary particle of mass m_2 , and let the velocity of the incident particle after the collision be v_1 and that of the struck particle be v_2 . The equation of conservation of energy is

$$\frac{1}{2}m_1v^2 = \frac{1}{2}m_1v_1^2 + \frac{1}{2}m_2v_2^2, \qquad (11-14)$$

.

:

and the equation of conservation of momentum is

$$m_1 v = m_1 v_1 + m_2 v_2. \tag{11-15}$$

The velocity v_1 can be eliminated from the last two equations, and a relationship is obtained between v_2 and v_1 ,

$$v_2 = \frac{2m_1}{m_1 + m_2} v. \tag{11-16}$$

If the incident particle is a neutron with known velocity v, its mass m_1 can be calculated from Eq. (11-16) if the mass m_2 and the maximum speed v_2 of the recoiling nucleus are known. Chadwick, however, did not have reliable information about the speed of the neutrons and had to use a less direct method. If a neutron with the same velocity v collides with a third particle of mass m_3 , the maximum velocity imparted to the latter is given by

$$v_3 = \frac{2m_1}{m_1 + m_3} v, \qquad (11-17)$$

where m_1 and v are the same as in Eq. (11-16). Upon dividing Eq. (11-16) by Eq. (11-17), we get

$$\frac{v_2}{v_3} = \frac{m_1 + m_3}{m_1 + m_2}$$
(11-18)

If m_2 , m_3 , v_2 , and v_3 are known, the mass m_1 of the neutron can be found. The maximum velocity imparted to hydrogen nuclei ejected from paraffin wax by the neutrons emitted by beryllium bombarded with polonium α -particles was found to be 3.3×10^9 cm/sec. The recoils of nitrogen nuclei after being struck by the same neutrons were observed in cloudchamber experiments and their maximum velocity was found to be 4.7×10^8 cm/sec. Taking m_2 and m_3 to be the masses of the proton and nitrogen nuclei, equal to 1 amu and 14 amu respectively, and with the corresponding values $v_2 = 3.3 \times 10^9$ and $v_3 = 4.7 \times 10^8$, we get from Eq. (11-18)

$$\frac{m_1+14}{m_1+1}=\frac{v_2}{v_3}=\frac{3.3 imes10^9}{4.7 imes10^8}$$
 ,

and $m_1 = 1.15$. This result, although approximate because of errors in the determination of the maximum recoil velocities, showed that the mass of the neutron is roughly the same as that of the proton.

Chadwick obtained a somewhat better value of the neutron mass by considering the reaction

$$_{5}B^{11} + _{2}He^{4} \rightarrow _{7}N^{14} + _{0}n^{1}$$
.

The Q-value was found from the kinetic energies of the incident α -particles, of the nitrogen nuclei (measured in cloud-chamber experiments), and of the neutron (estimated from the maximum recoil energy of a proton after being struck by a neutron). From the Q-value and the known atomic masses of B¹¹, He⁴, and N¹⁴, the mass of the neutron can be obtained with the aid of Eq. (11-6). In this way, Chadwick found a value between 1.005 and 1.008 atomic mass units. The best methods now available for determining the neutron mass give 1.008983 amu.

The proof of the existence of the neutron and the development of methods for analyzing nuclear reactions in which one of the products is a neutron led to the discovery of other reactions similar to that represented by Eq. (11-13). Among these are

$${}_{3}\text{Li}^{7} + {}_{2}\text{He}^{4} \rightarrow [{}_{5}\text{B}^{11}] \rightarrow {}_{5}\text{B}^{10} + {}_{0}\text{n}^{1},$$

$${}_{7}\text{N}^{14} + {}_{2}\text{He}^{4} \rightarrow [{}_{9}\text{F}^{18}] \rightarrow {}_{9}\text{F}^{17} + {}_{0}\text{n}^{1},$$

$${}_{11}\text{Na}^{23} + {}_{2}\text{He}^{4} \rightarrow [{}_{13}\text{Al}^{27}] \rightarrow {}_{13}\text{Al}^{26} + {}_{0}\text{n}^{1},$$

$${}_{13}\text{Al}^{27} + {}_{2}\text{He}^{4} \rightarrow [{}_{15}\text{P}^{31}] \rightarrow {}_{15}\text{P}^{30} + {}_{0}\text{n}^{1},$$

$${}_{18}\text{A}^{40} + {}_{2}\text{He}^{4} \rightarrow [{}_{20}\text{Ca}^{44}] \rightarrow {}_{20}\text{Ca}^{43} + {}_{0}\text{n}^{1}.$$

When these reactions are compared with those mentioned in Section 11-1, it is seen that some atoms, such as N¹⁴, Na²³, and A²⁷, emit either a proton or a neutron when bombarded with α -particles. The compound nucleus, e.g., [$_{9}F^{18}$], [$_{13}Al^{27}$], [$_{15}P^{31}$], may disintegrate by means of either process, giving a different product in the two cases. It will be seen in the next chapter that the products of these nuclear reactions are sometimes radioactive.

11-4 The acceleration of charged particles. The early artificial disintegrations were produced by bombarding materials with α -particles from natural radioactive substances. The neutrons from the bombardment of beryllium with α -particles were also used as projectiles and, as will be seen later, have produced many artificial disintegrations. The development of the field of nuclear transmutation was limited, however, because α -particles could be obtained only in beams of low intensity and with energies not greater than 7.68 Mev (RaC'), and transmutations by these particles were possible only with the lighter elements. Furthermore, it seemed possible that other particles, protons, deuterons, and even γ -rays, might yield interesting results when used as projectiles. Protons and deuterons with a single positive charge would experience smaller repulsive Coulomb forces than α -particles in the field of the nucleus, and it was thought that this effect might make disintegrations possible with lowenergy protons. It became important, therefore, to develop laboratory methods of accelerating charged particles to high energies, and several successful methods were invented about 1930.

Among the earliest particle accelerators were the voltage multiplier of Cockcroft and Walton, the electrostatic generator of Van de Graaff, and the cyclotron of Lawrence and Livingston. By the suitable choice of a machine and its operating conditions, protons, deuterons, or α -particles with energies as high as several Mev could be obtained by 1936. The further development of these machines and the design of new accelerators such as the frequency-modulated cyclotron and the synchrotron made it possible to obtain particles with energies up to hundreds of Mev. The "Cosmotron" at the Brookhaven National Laboratory has produced protons with energies of 2.5 billion electron volts, and recently built machines have raised the energies to 25 Bev. Electrons can be accelerated to energies up to 300 Mey in a betatron, or in a linear accelerator. When these electrons strike a metal target, highly energetic x-rays are produced, which can also be used as projectiles. Finally, the discovery of nuclear fission and the development of chain-reacting piles, or nuclear reactors (cf. Chapter 20), have supplied highly intense sources of neutrons for nuclear bombardment.

With the available particles and accelerators, a large variety of nuclear reactions can be made to occur. The rest of this chapter will be limited to the commonest reactions which take place with particle energies up to about 20 Mev. These reactions will serve to show the most important features of artificial disintegration, and the ideas involved can then be extended to the more complex reactions which occur with particle energies of the order of 100 Mev. The particle accelerators used to produce projectiles for nuclear reactions will be discussed in Chapter 21. For the purposes of the present chapter, it is enough to realize that various bombarding particles of almost any desired energy can be obtained with the help of suitable devices, and that these projectiles can cause many different kinds of nuclear reactions.

11-5 Transmutation by protons. The first case of a nuclear disintegration brought about entirely by artificial means was one for which protons were used as the projectiles. Cockcroft and Walton⁽⁵⁾ bombarded lithium with protons accelerated to energies of 0.1 to 0.7 Mev. Scintillations caused by particles ejected from the lithium were observed on a zinc sulfide screen placed a short distance away. The particles were proved to be α -particles by photographing their tracks in a cloud chamber,⁽⁶⁾ and the lithium isotope that was disintegrated was proved to be Li⁷. The cloud-chamber pictures showed that two α -particles leave the

$$_{3}\mathrm{Li}^{7} + {}_{1}\mathrm{H}^{1} \rightarrow [{}_{4}\mathrm{Be}^{8}] \rightarrow {}_{2}\mathrm{He}^{4} + {}_{2}\mathrm{He}^{4}.$$

This reaction has a certain historical interest because it provided one of the earliest quantitative proofs⁽⁷⁾ of the validity of the Einstein massenergy relationship. It was a good reaction for this purpose because the energies of the products could be measured precisely, and the masses were known. The proof will be repeated here, with newer values of the various quantities. The values of the atomic masses are, from Table 11-1,

$$M_X = M(\text{Li}^7) = 7.018222 \text{ amu},$$

 $m_x = m(\text{H}^1) = 1.008144,$
 $M_Y = m_y = m(\text{He}^4) = 4.003873.$

The value of Q obtained from the masses is $Q_m = 0.01860 \text{ amu} = 17.34 \text{ Mev}$. The best experimental value of Q, obtained from the energies of the incident protons and the emergent α -particles, is 17.33 Mev. This agreement shows clearly that the theoretical expression for Q, based on the mass-energy relationship, agrees with experiment, and that there was a genuine release of energy from the lithium atom at the expense of its mass. Since 1932, many nuclear transformations have been studied in detail and the results invariably agree with the relationships deduced from the Einstein equation.

The disintegration that has just been discussed is an example of the general type

$$_{Z}X^{A} + {}_{1}\mathrm{H}^{1} \rightarrow [_{Z+1}Cn^{A+1}] \rightarrow {}_{Z-1}Y^{A-3} + {}_{2}\mathrm{He}^{4}.$$
 (11-19)

Other examples are

$${}_{3}\text{Li}^{6} + {}_{1}\text{H}^{1} \rightarrow [{}_{4}\text{Be}^{7}] \rightarrow {}_{2}\text{He}^{3} + {}_{2}\text{He}^{4},$$

$${}_{4}\text{Be}^{9} + {}_{1}\text{H}^{1} \rightarrow [{}_{5}\text{B}^{10}] \rightarrow {}_{3}\text{Li}^{6} + {}_{2}\text{He}^{4},$$

$${}_{9}\text{F}^{19} + {}_{1}\text{H}^{1} \rightarrow [{}_{10}\text{Ne}^{20}] \rightarrow {}_{8}\text{O}^{16} + {}_{2}\text{He}^{4},$$

$${}_{13}\text{Al}^{27} + {}_{1}\text{H}^{1} \rightarrow [{}_{14}\text{Si}^{28}] \rightarrow {}_{12}\text{Mg}^{24} + {}_{2}\text{He}^{4}.$$

An interesting reaction occurs when the target is B^{11} ,

 $_{5}\mathrm{B}^{11} + _{1}\mathrm{H}^{1} \rightarrow [_{6}\mathrm{C}^{12}] \rightarrow _{4}\mathrm{Be}^{8} + _{2}\mathrm{He}^{4};$

 Be^8 is highly unstable and breaks up into two more α -particles,

$$_{4}\mathrm{Be}^{8} \rightarrow _{2}\mathrm{He}^{4} + _{2}\mathrm{He}^{4}$$
.

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The final result of the bombardment is three α -particles, and the reaction may be regarded as a case of multiple particle production.

Proton bombardment can also result in nuclear reactions in which one of the products is a neutron. The reaction is of the type

$$_{Z}X^{A} + {}_{1}\mathrm{H}^{1} \rightarrow [_{Z+1}Cn^{A+1}] \rightarrow _{Z+1}Y^{A} + {}_{0}n^{1}.$$
 (11-20)

The effect of the transmutation is to increase the charge on the nucleus by one unit, moving it one place to the right in the periodic table; the mass number is not changed. Examples of this reaction are

$${}_{5}B^{11} + {}_{1}H^{1} \rightarrow [{}_{6}C^{12}] \rightarrow {}_{6}C^{11} + {}_{0}n^{1},$$

$${}_{8}O^{18} + {}_{1}H^{1} \rightarrow [{}_{9}F^{19}] \rightarrow {}_{9}F^{18} + {}_{0}n^{1},$$

$${}_{28}Ni^{58} + {}_{1}H^{1} \rightarrow [{}_{29}Cu^{59}] \rightarrow {}_{29}Cu^{58} + {}_{0}n^{1}$$

In the (p,n) reactions, the mass change is usually negative, so that the reactions are endoergic. The reaction

$$_{29}\text{Cu}^{65} + _{1}\text{H}^{1} \rightarrow [_{30}\text{Zn}^{66}] \rightarrow _{30}\text{Zn}^{65} + _{0}\text{n}^{1}$$

is an example of a case for which the threshold energy has been measured.⁽⁸⁾ The source of the protons was an electrostatic generator, and the relative number of neutrons produced was determined as a function of the energy of the bombarding protons. The experimental results in the neighborhood of the threshold energy were then analyzed, and the value of the threshold energy was found to be $E_{\rm th} = 2.164 \pm 0.01$ Mev. The Q-value is obtained from Eq. (11-11), in which the masses M_X and m_x may be replaced by the mass numbers 65 and 1, respectively. Then

$$Q = -E_{\rm th} \times \frac{65}{66} = -2.164 \times \frac{65}{66} = -2.13$$
 Mev.

In some cases the bombarding proton is simply captured by a nucleus. The compound nucleus which is formed is again unstable, but becomes less unstable by emitting a γ -ray photon rather than a neutron or an α -particle. The reaction is of the type

$$_{Z}X^{A} + _{1}H^{1} \rightarrow [_{Z+1}Cn^{A+1}] \rightarrow _{Z+1}Y^{A+1} + \gamma,$$
 (11-21)

and some examples are

$${}_{3}\mathrm{Li}^{7} + {}_{1}\mathrm{H}^{1} \rightarrow [{}_{4}\mathrm{Be}^{8}] \rightarrow {}_{4}\mathrm{Be}^{8} + \gamma,$$

$${}_{6}\mathrm{C}^{12} + {}_{1}\mathrm{H}^{1} \rightarrow [{}_{7}\mathrm{N}^{13}] \rightarrow {}_{7}\mathrm{N}^{13} + \gamma,$$

$${}_{9}\mathrm{F}^{19} + {}_{1}\mathrm{H}^{1} \rightarrow [{}_{10}\mathrm{Ne}^{20}] \rightarrow {}_{10}\mathrm{Ne}^{20} + \gamma,$$

$${}_{13}\mathrm{Al}^{27} + {}_{1}\mathrm{H}^{1} \rightarrow [{}_{14}\mathrm{Si}^{28}] \rightarrow {}_{14}\mathrm{Si}^{28} + \gamma.$$

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The γ -ray photons emitted in these reactions are often very energetic and can, in turn, be used to produce nuclear disintegrations. The bombardment of lithium yields photons with an energy of 17.2 Mev, far more energetic than the 2.6-Mev photons which are the most energetic available from the natural radioactive nuclides.

Another type of disintegration caused by protons is that in which deuterons are produced. Examples of this reaction are

$${}_{4}\text{Be}^{9} + {}_{1}\text{H}^{1} \rightarrow [{}_{5}\text{B}^{10}] \rightarrow {}_{4}\text{Be}^{8} + {}_{1}\text{H}^{2},$$
$${}_{3}\text{Li}^{7} + {}_{1}\text{H}^{1} \rightarrow [{}_{4}\text{Be}^{8}] \rightarrow {}_{3}\text{Li}^{6} + {}_{1}\text{H}^{2}.$$

11-6 Transmutation by deuterons. A great many nuclear reactions have been observed with high energy deuterons as the bombarding particles. In most of these cases, the deuterons have been accelerated up to energies of several Mev in a cyclotron or in an electrostatic generator. One of the first deuteron-induced reactions studied was again that on a lithium atom,⁽⁹⁾

$$_{3}\text{Li}^{6} + {}_{1}\text{H}^{2} \rightarrow [{}_{4}\text{Be}^{8}] \rightarrow {}_{2}\text{He}^{4} + {}_{2}\text{He}^{4}.$$

Other examples of (d,α) reactions are

$${}_{8}O^{16} + {}_{1}H^2 \rightarrow [{}_{9}F^{18}] \rightarrow {}_{7}N^{14} + {}_{2}He^4,$$

 ${}_{13}Al^{27} + {}_{1}H^2 \rightarrow [{}_{14}Si^{29}] \rightarrow {}_{12}Mg^{25} + {}_{2}He^4.$

The mass change is usually positive so that the Q-values are positive, and the reactions are excergic. The general reaction of this type is

$$_{Z}X^{A} + _{1}H^{2} \rightarrow [_{Z+1}Cn^{A+2}] \rightarrow _{Z-1}Y^{A-2} + _{2}He^{4}.$$
 (11-22)

As in the case of proton bombardment, disintegrations produced by deuterons do not always yield α -particles. Deuteron-proton reactions are often found, such as

$$_{6}C^{12} + _{1}H^{2} \rightarrow [_{7}N^{14}] \rightarrow _{6}C^{13} + _{1}H^{1},$$

 $_{11}Na^{23} + _{1}H^{2} \rightarrow [_{12}Mg^{25}] \rightarrow _{11}Na^{24} + _{1}H^{1},$
 $_{15}P^{31} + _{1}H^{2} \rightarrow [_{16}S^{33}] \rightarrow _{15}P^{32} + _{1}H^{1}.$

The result of these transformations is to increase the mass of the nucleus by one unit, leaving the charge unchanged. The general (d,p) reaction is

$$_{Z}X^{A} + {}_{1}\mathrm{H}^{2} \rightarrow [_{Z+1}Cn^{A+2}] \rightarrow _{Z}X^{A+1} + {}_{1}\mathrm{H}^{1}.$$
 (11-23)

The Q-values for these reactions are usually positive, so that the reactions are excergic.

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Neutrons are often produced as a result of deuteron bombardment, as in the following reactions:

$${}_{6}C^{12} + {}_{1}H^{2} \rightarrow [{}_{7}N^{14}] \rightarrow {}_{7}N^{13} + {}_{0}n^{1},$$

$${}_{4}Be^{9} + {}_{1}H^{2} \rightarrow [{}_{5}B^{11}] \rightarrow {}_{5}B^{10} + {}_{0}n^{1},$$

$${}_{3}Li^{7} + {}_{1}H^{2} \rightarrow [{}_{4}Be^{9}] \rightarrow {}_{4}Be^{8} + {}_{0}n^{1}.$$

The general (d,n) reaction is represented by the equation

$$_{Z}X^{A} + {}_{1}\mathrm{H}^{2} \rightarrow [_{Z+1}Cn^{A+2}] \rightarrow _{Z+1}Y^{A+1} + {}_{0}\mathrm{n}^{1}.$$
 (11-24)

One of the most interesting cases of deuteron bombardment is that in which the target contains deuterons. Deuterium targets have been made by freezing deuterium oxide $(D_2O-$ "heavy water") onto a surface kept cold by liquid air. Both the (d,p) and the (d,n) reactions have been observed,

$$_{1}H^{2} + _{1}H^{2} \rightarrow [_{2}He^{4}] \rightarrow _{1}H^{3} + _{1}H^{1},$$

 $_{1}H^{2} + _{1}H^{2} \rightarrow [_{2}He^{4}] \rightarrow _{2}He^{3} + _{0}n^{1}.$

The excited compound nucleus $[_2\text{He}^4]$ can disintegrate in two ways. In the first, a proton and a new isotope of hydrogen are formed; in the second, a neutron and an isotope of helium are formed. The new hydrogen isotope has a mass very nearly equal to 3 atomic mass units, and has been given the name tritium. It is unstable and has a half-life of about 12 years. The helium isotope of mass 3 is stable and is found in nature.

11-7 Transmutation by neutrons. Neutrons have proved to be especially effective in producing nuclear transformations. Since they have no electric charge, they are not subject to repulsive electrostatic forces in the neighborhood of a positively charged nucleus, and are therefore more likely to penetrate nuclei than are protons, deuterons, or α -particles. Not only are highly energetic neutrons capable of causing nuclear reactions. but slowly moving neutrons are also extremely effective. Because of these properties of the neutron many more disintegrations have been produced with neutrons than with any other particle. Before the development of the chain-reacting pile, or nuclear reactor, the main sources of neutrons were reactions such as $H^{2}(d,n)He^{3}$, $Be^{9}(d,n)B^{10}$ and $Be^{9}(\alpha,n)C^{12}$. The fast neutrons produced were slowed down by allowing them to pass through some hydrogen-containing substance such as water or paraffin. A neutron gives up a large fraction of its energy in a collision with a hydrogen nucleus and, after many collisions, the average energy of the neutrons is reduced to a few hundredths of an electron volt. These slow neutrons have been found to be especially useful from a practical standpoint, and their interactions with nuclei are particularly interesting.

The reaction between a neutron and a nucleus gives rise in most cases to an α -particle, a proton, a γ -ray photon, or to two neutrons. The (n,α) reaction⁽¹⁰⁾ is represented by the general equation

$$_{Z}X^{A} + _{0}n^{1} \rightarrow [_{Z}Cn^{A+1}] \rightarrow _{Z-2}Y^{A-3} + _{2}\text{He}^{4}.$$
 (11-25)

Among the most interesting examples of this reaction are

$${}_{3}\text{Li}^{6} + {}_{0}n^{1} \rightarrow [{}_{3}\text{Li}^{7}] \rightarrow {}_{1}\text{H}^{3} + {}_{2}\text{He}^{4},$$

 ${}_{5}\text{B}^{10} + {}_{0}n^{1} \rightarrow [{}_{5}\text{B}^{11}] \rightarrow {}_{3}\text{Li}^{7} + {}_{2}\text{He}^{4},$
 ${}_{13}\text{Al}^{27} + {}_{0}n^{1} \rightarrow [{}_{13}\text{Al}^{28}] \rightarrow {}_{11}\text{Na}^{24} + {}_{2}\text{He}^{4}.$

The first two reactions have a relatively high yield and are therefore often used to detect neutrons. In one method, an ionization chamber is lined with boron, usually in the form of a compound. The capture of a neutron by an atom of the B¹⁰ isotope causes the liberation of an α -particle, which is detected by the ionization it produces in the chamber.

In some cases, the compound nucleus formed by the capture of a neutron emits a proton. The (n,p) reaction is described by the equation

$$_{Z}X^{A} + _{0}n^{1} \rightarrow [_{Z}Cn^{A+1}] \rightarrow _{Z-1}Y^{A} + _{1}H^{1}.$$
 (11-26)

The effect of this reaction is to replace a proton in the nucleus by a neutron; the mass number is not changed, but the charge is decreased by one unit and the atom is moved one place to the left in the periodic table. Some examples of this reaction are

$$_{7}N^{14} + _{0}n^{1} \rightarrow [_{7}N^{15}] \rightarrow _{6}C^{14} + _{1}H^{1},$$

 $_{13}Al^{27} + _{0}n^{1} \rightarrow [_{13}Al^{28}] \rightarrow _{12}Mg^{27} + _{1}H^{1}.$

The first of these reactions can be induced by slow neutrons; with heavier nuclei, more energetic neutrons must be used, as in the case of Al^{27} and in the reaction

$$_{30}$$
Zn⁶⁴ + $_{.0}$ n¹ \rightarrow [$_{30}$ Zn⁶⁵] \rightarrow $_{29}$ Cu⁶⁴ + $_{1}$ H¹.

In another type of reaction, one neutron is captured by the nucleus and two neutrons are emitted. The (n,2n) reaction leaves the charge of the nucleus unchanged and decreases the mass number by one unit. The result is an isotope of the target nucleus with a mass number one unit smaller,

$$_{Z}X^{A} + _{0}n^{1} \rightarrow [_{Z}Cn^{A+1}] \rightarrow _{Z}X^{A-1} + _{0}n^{1} + _{0}n^{1}.$$
 (11-27)

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An example of this reaction is

$$_{13}\mathrm{Al}^{27} + _{0}\mathrm{n}^{1} \rightarrow [_{13}\mathrm{Al}^{28}] \rightarrow _{13}\mathrm{Al}^{26} + _{0}\mathrm{n}^{1} + _{0}\mathrm{n}^{1}.$$

The mass change in the (n,2n) reaction is always negative; the Q-value is negative and fast neutrons are needed to bring about this reaction.

The commonest process which results from neutron capture is *radiative* capture.⁽¹¹⁾ represented by

$$_{Z}X^{A} + _{0}n^{1} \rightarrow [_{Z}Cn^{A+1}] \rightarrow _{Z}X^{A+1} + \gamma.$$
 (11-28)

The compound nucleus emits one or more γ-ray photons, and the final nucleus is an isotope of the target nucleus with a mass number one unit greater. The (n,γ) process has been observed in nearly all of the elements. The Q-values are always positive, the excess energy being carried away by the γ-rays. The simplest (n,γ) reaction with slow neutrons occurs with hydrogen as the target nucleus,

$$_{1}\mathrm{H}^{1} + _{0}\mathrm{n}^{1} \rightarrow [_{1}\mathrm{H}^{2}] \rightarrow _{1}\mathrm{H}^{2} + \gamma.$$

The product of the reaction is deuterium. When deuterium is bombarded with slow neutrons, tritium is formed,

$$_{1}\mathrm{H}^{2} + _{0}\mathrm{n}^{1} \rightarrow [_{1}\mathrm{H}^{3}] \rightarrow _{1}\mathrm{H}^{3} + \gamma.$$

Other typical (n, γ) reactions are

$${}_{13}\mathrm{Al}^{27} + {}_{0}\mathrm{n}^{1} \rightarrow [{}_{13}\mathrm{Al}^{28}] \rightarrow {}_{13}\mathrm{Al}^{28} + \gamma,$$

$${}_{49}\mathrm{In}^{115} + {}_{0}\mathrm{n}^{1} \rightarrow [{}_{49}\mathrm{In}^{116}] \rightarrow {}_{49}\mathrm{In}^{116} + \gamma,$$

$${}_{92}\mathrm{U}^{238} + {}_{0}\mathrm{n}^{1} \rightarrow [{}_{92}\mathrm{U}^{239}] \rightarrow {}_{92}\mathrm{U}^{239} + \gamma.$$

The radiative capture of slow neutrons often results in product nuclei which are radioactive, and this reaction is one of the most important sources of artificial radioactive nuclides.

11-8 Transmutation by photons. Atomic nuclei can also be disintegrated by bombardment with high-energy photons, a process which is usually called *photodisintegration*. Since the photon has no mass, it can supply only its kinetic energy to a nuclear reaction. This energy must be at least as great as the binding energy of a nuclear particle before such a particle can be ejected from a nucleus. Photodisintegration reactions are, therefore, endoergic and usually have threshold energies of the order of 10 Mev. With only two exceptions, photodisintegration does not occur with γ -rays from natural radioactive substances. These exceptions are the deuteron, which has a binding energy of only 2.2 Mev, and the nuclide 4Be⁹,

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in which one neutron is loosely bound. In the case of the deuteron, $^{(12)}$ the reaction is

$$_{1}\mathrm{H}^{2} + \gamma \rightarrow [_{1}\mathrm{H}^{2}] \rightarrow _{1}\mathrm{H}^{1} + _{0}\mathrm{n}^{1},$$

and is the reverse of the radiative capture of a neutron by a proton. In beryllium, the reaction is

$$_{4}\text{Be}^{9} + \gamma \rightarrow [_{4}\text{Be}^{9}] \rightarrow _{4}\text{Be}^{8} + _{0}\text{n}^{1},$$

with a Q-value of -1.67 Mev.

It will be recalled from Section 11-5 that when Li is bombarded with protons, 17-Mev γ -rays are produced. These have been used successfully to cause the photodisintegration of other nuclides, as in the (γ, n) reaction

$$_{15}P^{31} + \gamma \rightarrow [_{15}P^{31}] \rightarrow _{15}P^{30} + _{0}n^{1}$$

The (γ, p) reaction requires still higher energies, and has been observed with high-energy photons from a betatron.

21.4 Archaeological Dating by the 14C Method

Carbon has three important isotopes, 12, 13 and 14 having terrestrial abundances of 98.89% m 1.11% and zero, respectively. Of these, ¹⁴C is unstable and decays according to the equation

$${}^{4}C \rightarrow {}^{14}\gamma N + {}^{0}_{-1}e(\beta^{-}) + \tilde{\nu}$$

with $T_{1/2} = 5730$ a and $E_{\beta^-} = 0.158$ MeV.

There is a trace of ¹⁴C in the atmosphere due to cosmic neutron bombardment of 14N, thus

$${}^{14}_{7}N + {}^{1}_{0}n \rightarrow {}^{14}_{6}C + {}^{1}_{1}H(p).$$

If these two equations are taken together it seems reasonable to assume that over, say, ten half-lives of ¹⁴₆C decay the quantity of ¹⁴CO₂ present in the atmosphere has always been constant. In the atmosphere, therefore, the formation and decay of ¹⁴C are in equilibrium. By a similar argument we can assume that the concentration of ¹⁴C in all living vegetable tissue is the same, due to the fact that the carbon dioxide taken in by the plants from the atmosphere contains this constant quantity of ¹⁴CO₂. However, when the plant dies and no longer takes in CO2 from the atmosphere, the 14C equilibrium quantity attained during the life of the plant now begins to decay by β^- -emission with the half-life of 5730 ± 30 a.

Suppose a sample of this dead matter (wood, charcoal, book-binding, peat, rope, etc.) is now measured, say t years later. With the usu

$$N = N_0 e^{-\lambda t}$$

or

$$\ln N = \ln N_0 - \lambda t$$

Since

$$\lambda = \frac{0.6931}{T_{1/2}},$$

we get

$$\log N = \log N_0 - 0.3010 t/T_{1/2}$$

where N_0 is the activity at death and N is the activity at present, referring to the β -emission of the sample with $T_{1/2} = 5730$ years. Thus if N_0 is the original count rate, it is also the present count rate of the β^- -emission from living matter i.e. the equilibrium activity previously mentioned. This value is about $16\cdot 1 + 0\cdot 3$

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counts per minute per gram of carbon. In order to measure the value of N with any degree of accuracy one must know the efficiency of the counter and the background count with an accuracy of about 2%. When this is done, and using the value of $T_{1/2}$ previously stated, the value of t or the age of the specimen can be found.

Two interesting ages will be mentioned here, one obtained from the charcoal from Stonehenge, England, and the other from the binding of the Isaiah scroll in the Dead Sea scrolls. By the ¹⁴C method Stonehenge is found to be 3798 ± 275 years old and the Dead Sea scroll 1917 ± 200 years old. The errors quoted reflect the difficulties associated with ¹⁴C dating.

The whole method rests on the assumptions that the ¹⁴C content of the atmosphere has been constant over the last 50 000 years, i.e. the cosmic ray intensity over this period has not varied. The ¹⁴C method also takes for granted a constancy of ¹⁴N over the same period. Finally it must be assumed that there has been no secondary interference during the decay period and the sample has remained the same since its 'death', except for the β^- -decay mechanism.

Chapter 2

NUCLEAR REACTIONS

In the discussion of nuclear reactions in Chapter 11, the emphasis was on the production of new nuclear species and the interest was mainly in the change of one nucleus into another. The quantitative aspects stressed were those of the mass and energy balance. The study of nuclear reactions is also important for other reasons. Information about the relative probabilities of different reactions provides clues to the problem of nuclear structure and offers a testing ground for ideas about nuclear forces. Any nuclear transmutation process may lead to an excited state of the product nucleus, and the decay of the excited state then gives information about energy levels and decay schemes. The process may be caused by any kind of incident particle and may result in the emission of any kind of particle (proton, neutron, deuteron, α -particle, or γ -ray). The incident and emitted particle may even be of the same kind, as in scattering processes. The great number of nuclear reactions which are possible provide, therefore, a wealth of experimental data for the field of nuclear spectroscopy and for the theory of nuclear structure. It is from this standpoint that nuclear reactions will be considered in the present chapter.

16-1 Nuclear reactions and excited states of nuclei. In Chapter 11, the nuclear reactions considered were of the type

$$x + X \to Y + y, \tag{16-1}$$

or, in more compact notation, X(x, y)Y. The equation and the notation both mean that particle x strikes nucleus X to produce nucleus Y and particle y. The particles x and y may be elementary particles or γ -rays, or they may themselves be nuclei, e.g., α -particles or deuterons. The transmutation represented by Eq. (16-1) does not cover all the reactions of interest. In the general case, more than one particle may emerge, or the outgoing particle may be the same as the incident particle. For the purpose of this chapter, the nuclear reactions to be considered may be represented by the set of equations

$$x + X \to \begin{cases} X + x, \\ X^* + x, \\ Y + y, \\ Z + z, \text{ etc.} \end{cases}$$
(16-2)

In the first two reactions of the set, the outgoing particle is of the same kind as the incident particle, and the process is called scattering. The first reaction represents *elastic scattering*, in which the total kinetic energy of the system, projectile plus target, is the same before the collision as after. Some kinetic energy is usually transferred from the projectile to the target nucleus, but the latter is left in the same internal, or nuclear. state as before the collision. A collision between two billiard balls, in which neither ball is damaged or otherwise changed, is an example of an elastic collision. The second reaction represents inelastic scattering, in which the target nucleus X is raised into an excited state X^* , and the total kinetic energy of the system is decreased by the amount of the excitation energy given to the target nucleus. The other reactions of the set represent different possible nuclear transmutations in which the product nuclei may be formed in their ground states or, more often, in excited states. The excited product nucleus usually decays very quickly to the ground state with the emission of γ -rays.

The fact that the product nucleus in a transmutation process can be left in an excited state was discovered by measuring the energies of the protons emitted in (α, p) reactions on light elements, the first nuclear reactions studied in detail. When a given light element was bombarded with monoenergetic natural α -particles, one or more groups of protons, each containing particles of the same energy, was observed for a particular direction of emission.^(1,2) The existence of the distinct proton groups was demonstrated by their different ranges. When boron was bombarded with α -particles from polonium with a range of 3.8 cm, or an energy of 5.30 Mev, two groups of emergent protons were found at right angles to the incident α -particle beam; these protons had ranges of 20 cm and 50 cm, respectively. Similar results were obtained with other light elements such as fluorine and aluminum; when the latter was bombarded with α -particles from RaC', four groups of protons were observed⁽³⁾ for each of several different energies of the incident α -particles.

The energy balance of the reactions, or Q-value, can be calculated for each energy group by the methods of Section 11-2. The proton group with the greatest energy gives the greatest Q-value, and this value is supposed to correspond to the ground state of the product nucleus. A proton group of lower energy gives a lower Q-value, and the difference between the greatest Q and a smaller one gives the excitation energy of the product nucleus after emission of the lower energy proton. These ideas and their application involve some of the basic procedures of nuclear physics and will be illustrated by an analysis of two experiments, on the reaction

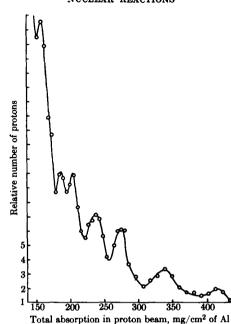
$$_{13}\text{Al}^{27} + _{2}\text{He}^{4} \rightarrow [_{15}\text{P}^{31}] \rightarrow _{14}\text{Si}^{30} + _{1}\text{H}^{1} + Q,$$

which are representative of work relating nuclear reactions to energy levels. In one experiment,⁽⁴⁾ an aluminum foil was bombarded with α -particles accelerated in a cyclotron to an energy of 7.3 Mev. The ranges of the protons emerging at an angle of 0° with the direction of the incident beam were measured by counting coincidences between protons and γ -rays. Four groups of protons were observed with ranges in air of 101.6 cm, 60.8 cm. 40.8 cm, and 25-30 cm, respectively. The corresponding energies, obtained from range-energy curves for protons, are 9.34, 6.98, 5.55, and 4.2-4.65 Mev. The energies of the incoming α -particle and the outgoing proton are known for each group, as are the mass numbers of the initial and final nuclei and, since the angle at which the protons are observed is 0° , Q-values can be calculated from Eq. (11-10). The greatest proton energy gives Q = 2.22 Mev, which corresponds to the formation of Si³⁰ in its ground state; the three lower-energy proton groups give Q-values of -0.06, -1.44, and about -2.4 MeV, respectively. There are, therefore, three excited states of Si³⁰ with energies 2.28, 3.66, and about 4.6 Mev above that of the ground state. The ground state energy is taken as the zero point of the energy scale for the various states. The results, which are collected in the first part of Table 16-1, are in good agreement with those of earlier experiments.^(3,5) It was also found in this experiment

TABLE 16-1

Experiment $1^{(4)}$ Energy of α -particles = 7.3 Mev			Experiment $\mathcal{E}^{(6)}$ Energy of α -particles = 21.54 Mev		
Energy of protons emitted at 0°, Mev	Q-value, Mev	Energy of excited state, Mev	Energy of protons emitted at 90°, Mev	Q-value, Mev	Energy of excited state, Mev
9.34 6.98	2.22 0.06	0 2.28		2.22	0
5.55	1.44	3.66	16.85	-1.27	3.49
4.2-4.65	-2.4	4.6	14.96	-3.22	5.44
			13.28	-4.96	7.18
			12.29 11.27		8.20 9.26
			10.68	-7.65	9.87
			9.72	-8.64	10.86

Excited States of Si³⁰ from the Bombardment of Aluminum by Alpha-Particles





that no γ -rays accompanied the highest-energy group of protons, while the lower-energy protons did show coincidences with γ -rays; the lowerenergy protons are indeed associated with excited states of the Si³⁰ nucleus, while the highest-energy group is associated with the ground state.

In another experiment,⁽⁶⁾ a thin aluminum foil was bombarded in vacuum with α -particles accelerated in a cyclotron to an energy of 21.54 Mev. The protons emerging at an angle of 90° with the direction of the incident beam were counted with two proportional counters placed in series and arranged for coincidence counting. The ranges of the proton groups in aluminum were measured by allowing the protons to pass through aluminum foils before entering the counting system. Under the conditions of the counting procedure, the number of protons counted in a particular group showed a peak at the end of the range so that when the number of counts is plotted against the thickness of aluminum, each peak in the curve corresponds to the range or energy of a different proton group. The change of the counting rate with the thickness of the aluminum absorber is shown in Fig. 16-1. The energies of the proton groups were obtained from the ranges in aluminum by means of the known theoretical relationship between range and energy in that material, and are listed in the second part of Table 16-1. The Q-values are obtained from Eq. (11-9). The range of the most energetic proton group could not be determined well in this experiment because the counting rates became too low, so the value Q = 2.22 Mev was used for the reaction leading to the ground state of Si³⁰, and gave the energy levels listed in the table. The energy level values at 3.66 Mev and 3.49 Mev correspond to the same excited state, the discrepancy being experimental. The level at about 4.6 Mev observed in the first experiment was not observed in the second experiment, but other work has established the existence of a level at 4.75 Mev. There are, therefore, at least 9 excited energy levels of Si³⁰, as shown in the table, and it is probable that there are more which have not been excited by the (α, p) reaction.

The emission of groups of particles is not limited to (α, p) reactions, but is also found in many other reactions. In each case, the different energy groups correspond to different states of the product nucleus, the lower-energy groups corresponding to excited states which decay to the ground state by emitting γ -rays.

Another important feature of many nuclear reactions was discovered during the investigation of (α, p) reactions when the energy of the incident α -particles was varied. It was found in the early experiments that when the energy of the incident α -particles was increased the yield of protons did not increase monotonically, as might be expected on intuitive grounds, but showed sharp maxima at certain discrete values of the energy.⁽³⁾ In the bombardment of Al by α -particles, the yield of protons had peaks at α -particle energies of 4.0, 4.49, 4.86, 5.25, 5.75, and 6.61 Mev, and was markedly lower in the energy range between any two of these peaks. The occurrence of maxima in the reaction rate at different energies is called resonance, and the particular energies at which the maxima occur are called resonance energies. The reason for the use of these terms will be discussed later. At each resonance energy of the incident α -particles the different proton groups are observed, and Q-values can be calculated from the known resonance energy and the corresponding energies of the proton groups. These Q-values are the same as those obtained for nonresonance α -particle energies, and lead to the same excited states of the product nucleus. The experimental results obtained at the resonance energies are often more useful than those obtained at other energies because more events occur at the resonance energies and more precise information can be obtained about the Q-values and excited levels in the compound nucleus. The resonance phenomenon does not, however, add anything new about the energy levels of the product nucleus.

Resonance is important in nuclear reactions because knowledge of the resonance energies in a reaction yields information about certain energy levels of the nucleus referred to in Chapter 11 as the *compound nucleus*.

NUCLEAR REACTIONS

The transmutations discussed in Chapter 11 were written in a way which indicates that the incident particle combines with the target nucleus to form an intermediate compound nucleus. When Al is bombarded with α -particles, the compound nucleus is $[P^{31}]$, and it turns out that the existence of resonance energies is related to the energy levels of P^{31} . Resonance is not limited to (α, p) reactions but is also found in other nuclear reactions.

16-2 The compound nucleus. In 1936, Bohr proposed his theory^(7,8) of the compound nucleus, which has been extremely useful in the correlation and interpretation of nuclear reactions. The basic ideas of this theory will therefore be considered before a detailed discussion of nuclear reactions is undertaken. Bohr assumed that a nuclear reaction takes place in two steps:

(1) The incident particle is absorbed by the initial, or target, nucleus to form a *compound nucleus*.

(2) The compound nucleus disintegrates by ejecting a particle (proton, neutron, α -particle, etc.) or a γ -ray, leaving the final, or product, nucleus.

Bohr assumed also that the mode of disintegration of the compound nucleus is independent of the way in which the latter is formed, and depends only on the properties of the compound nucleus itself, such as its energy and angular momentum. The two steps of the reaction can then be considered separate processes:

1. Incident particle + initial nucleus \rightarrow compound nucleus.

2. Compound nucleus \rightarrow product nucleus + outgoing particle.

Bohr's assumptions are in accord with many of the facts of nuclear transmutation. It was shown in Section 11-9 that when a given nuclide is bombarded with particles of a single type, several different new nuclides can be formed. When Al²⁷ is bombarded with protons, the new nuclide may be Mg²⁴, Si²⁷, Si²⁸, or Na²⁴. According to Bohr, the interaction between Al²⁷ and a proton is assumed to give the compound nucleus [14Si²⁸], which can then disintegrate in any one of several ways: into Mg²⁴ and an α -particle; or into Si²⁷ and a neutron; or into Si²⁸ and a γ -ray; or even into Na²⁴, 3 protons, and a neutron. Bohr's assumption is also in accord with the picture of the nucleus as a system of particles held together by very strong short-range forces. When the incident particle enters the nucleus, its energy is quickly shared among the nuclear particles before re-emission can occur, and the state of the compound nucleus is then independent of the way it was formed. That this conclusion is reasonable can be shown by the following arguments. On being captured, the incident particle makes available a certain amount of excitation energy. which is nearly equal to the kinetic energy of the captured particle plus its binding energy in the compound nucleus. The magnitude of the excitation energy can be calculated from the masses of the incident particle, target nucleus, and compound nucleus, and the kinetic energy of the incident particle. Consider, for example, the capture of a neutron by Al^{27} to form the compound nucleus $[Al^{28}]$,

 $_{13}\text{Al}^{27} + _{0}\text{n}^{1} \rightarrow [_{13}\text{Al}^{28}].$

The masses of the interacting neutron and nucleus are 1.00898 and 26.99008 amu, or a total of 27,99906 amu; that of the compound nucleus is 27,99077 amu. The mass excess is 0.00829 amu, corresponding to 7.72 Mev. to which must be added the kinetic energy of the incident neutron. In the case of a slow neutron, the kinetic energy may be neglected. If the incident neutron has a kinetic energy of 1 Mey, the excitation energy is nearly 7.72 + 1 = 8.72 Mev and the energy of the compound nucleus $[_{13}Al^{28}]$ is greater than the energy of the ground state of Al²⁸ by this amount. Immediately after the formation of the compound nucleus, the excitation energy may be considered to be concentrated on the captured particle, but this additional energy is rapidly distributed among the other particles in the compound nucleus as a result of interactions among the nuclear particles. The distribution presumably takes place in a random way. At a given instant, the excitation energy may be shared among several nucleons; at a later time it may be shared by other nucleons, or it may eventually again become concentrated on one nucleon or combination of nucleons. In the latter case, if the excitation energy is large enough, one nucleon, or a combination of nucleons, may escape, and the compound nucleus disintegrates into the product nucleus and outgoing particle. The energy that must be concentrated on a single nuclear particle or group of particles in order to separate it from the compound nucleus is called the separation or dissociation energy, and is usually about 8 Mev.

As a result of the random way in which the excitation energy is distributed in the compound nucleus, the latter has a lifetime which is relatively long compared with the time that would be required for a particle to travel across the nucleus. The latter time interval, sometimes called the *natural nuclear time*, is of the order of magnitude of the diameter of the nucleus divided by the speed of the incident particle. If the incident particle is a 1-Mev neutron, its speed is about 10^9 cm/sec. Since the diameter of the nucleus is of the order of 10^{-12} cm, the time required for a 1-Mev neutron to cross the nucleus is of the order of 10^{-21} sec. Even a slow neutron with a velocity of 10^5 cm/sec would need only about 10^{-17} sec to cross the nucleus. It will be seen below that the lifetime of a compound nucleus may be as long as 10^{-15} or 10^{-14} sec, which is long compared with the natural nuclear time. During its relatively long lifetime, the compound nucleus "forgets" how it was formed, and the disintegration

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is independent of the mode of formation. The compound nucleus may be said to exist in a "quasi-stationary" state, which means that although it exists for a time interval which is very long compared with the natural nuclear time, it can still disintegrate by ejecting one or more nucleons. These quasi-stationary states are usually called *virtual states* or *virtual levels* in contrast to *bound states* or *bound levels*, which can decay only by emitting γ -radiation.

There are many ways in which the excitation energy of the compound nucleus can be divided among the nuclear particles and, since each distribution corresponds to a virtual level, there are many possible virtual levels of the compound nucleus. These levels are closely related to the phenomenon of resonance. It is reasonable to assume that if the energy of the incident particle is such that the total energy of the system, incident particle plus target nucleus, is equal to the excitation energy of one of the levels of the compound nucleus, the probability that the compound nucleus will be formed is much greater than if the energy falls in the region between two levels. The system is analogous to that of a radio wave and a tuned receiver circuit. When the frequency (energy) of the incoming wave is equal to that of the circuit, the wave and circuit are in resonance and the reception is good; when the two frequencies (energies) are not equal, the reception is poor. The occurrence of a resonance peak in the rate of a nuclear reaction when the energy of the incoming particles is varied shows that the compound nucleus has an energy level whose excitation energy is very nearly the sum of the binding energy of that particle and its kinetic energy. In the Al²⁷(α , p)Si³⁰ reaction discussed above, 18 resonances have been observed at α -particle energies from 3.95 Mev to 8.62 Mev; the binding energy of the α -particle in the compound nucleus $[P^{31}]$ is 9.68 Mev. The excitation energy of a level cannot be obtained just by adding the resonance energy to the binding energy, because that would neglect the motion of the compound nucleus. Some of the energy of the incident particle is used in supplying kinetic energy to the compound nucleus and is not available for excitation.

The fraction of the particle energy that must be added to the binding energy in order to get the excitation energy can be derived as follows. If the target nucleus is assumed to be at rest before the collision, the momentum of the incident particle must be equal to that of the compound nuccleus,

$$m_x v = M_{CN} V,$$

where v, V, and M_{CN} are the speed of the incident particle, and the speed and mass of the compound nucleus, respectively. Then

$$V = v \, \frac{m_x}{M_{CN}} \cdot$$

The portion of the energy of motion of the incident particle that goes into exciting the initial nucleus will be called E'_{z} , and is given by

$$E'_{x} = \frac{1}{2} m_{x} v^{2} - \frac{1}{2} M_{CN} V^{2} = \frac{1}{2} m_{x} v^{2} - \frac{1}{2} M_{CN} v^{2} \frac{m_{x}^{2}}{M_{CN}^{2}}$$

$$= \frac{1}{2} m_{x} v^{2} \left(1 - \frac{m_{x}}{M_{CN}} \right) = E_{x} \left(1 - \frac{m_{x}}{M_{CN}} \right).$$
(16-3)

In calculations of this kind, the masses may be replaced by mass numbers without introducing any serious errors. Equation (16-3) may also be written

$$E'_x = E_x \left(\frac{M_X}{m_x + M_X} \right), \tag{16-4}$$

since $M_{CN} = M_X + m_x$.

The energies of the excited levels of $[P^{31}]$ which correspond to α -particle resonances are given in Table 16-2. The excitation energies of these levels are considerably greater than the dissociation energy (~8 Mev) for a particle and the compound nucleus can disintegrate by particle emission so that the levels are virtual. The reactions $Al^{27}(\alpha, p)Si^{30}$ and $Al^{27}(\alpha, n)P^{30}$ have been observed, a result which is consistent with the idea that the compound nucleus $[P^{31}]$ formed by the bombardment of aluminum with α -particles can disintegrate in more than one way.

Each excited state of the compound nucleus, whether bound or virtual, has a certain mean lifetime τ ; there is a certain period of time, on the average, during which the nucleus remains in a given excited state before decaying by emission of either a particle or a γ -ray. The reciprocal of the mean life is the disintegration constant, which gives the probability per unit time of the emission of a particle or γ -ray. In the discussion of energy states excited by nuclear reactions it is customary to use, instead of the disintegration constant, a quantity proportional to it, called the *level width*, and defined by the relation

$$\Gamma = \frac{h}{2\pi\tau} \,. \tag{16-5}$$

The level width has the units of energy, and its use is based on an application of the Heisenberg uncertainty principle (Section 7-8). According to this principle, the accuracy with which the energy and time can be determined for a quantized system such as an atomic nucleus is limited by the relationship

$$(\Delta E) \ (\Delta t) \ \sim \frac{h}{2\pi}, \qquad (16-6)$$

TABLE 16–2

Levels of the Compound Nucleus $[P^{31}]$ Excited in the Bombardment of Aluminum by α -Particles

α-particle resonance energy, Mev	Energy to be added to the binding energy, Mev	Excited level of [P ³¹], Mev	
3.95	3.44	13.12	
4.53	3.95	13.63	
4.70	4.09	13.77	
4.84	4.22	13.90	
5.12	4.46	14.14	
5.29	4.61	14.29	
5.64	4.91	14.59	
6.01	5.23	14.91	
6.38	5.56	15.24	
6.57	5.72	15.40	
7.00	6.10	15.78	
7.20	6.27	15.95	
7.34	6.39	16.07	
7.60	6.62	16.30	
8.04	7.00	16.68	
8.24	7.18	16.86	
8.42	7.33	17.01	
8.62	7.51	17.19	

where ΔE is the uncertainty in the energy and Δt is the uncertainty in the time. The mean lifetime of an excited state may be identified with the uncertainty Δt corresponding to an uncertainty ΔE in the energy. The latter is defined as the width Γ , in energy, of the excited level, giving Eq. (16-5). A state with a very short mean lifetime is poorly defined in energy and the width Γ is relatively large, while a long-lived state is sharply defined in energy and the width is relatively small. For each possible mode of decay, there is a different probability of decay and, therefore, a different *partial width* Γ_y for each decay product. The total width Γ of an energy level is then the sum of the individual partial widths;

$$\Gamma = \Gamma_{\gamma} + \Gamma_{\alpha} + \Gamma_{p} + \cdots, \qquad (16-7)$$

where Γ_{γ} , the radiation width, is a measure of the probability per unit time for emission of a γ -ray, Γ_{α} is the *a*-particle width, Γ_{p} is the proton width, and so on.

The concept of level widths is useful because values of these widths can often be obtained from measurements of resonances, as will be shown later. When the total width is known, the value of the mean lifetime is given by

$$\tau (\text{sec}) = \frac{h (\text{erg-sec})}{2\pi\Gamma (\text{erg})} = \frac{1.06 \times 10^{-27}}{\Gamma (\text{ev}) \times 1.60 \times 10^{-12}} = \frac{6.6 \times 10^{-16}}{\Gamma (\text{ev})} \cdot$$
(16-8)

Level widths are usually given in ev; for a wide level, Γ may be of the order of 10^4 ev, as in the case of light nuclei, and the lifetime is 6.6×10^{-20} sec, while a sharp level with a Γ of 0.1 ev has the relatively long mean life of 6.6×10^{-15} sec. When the narrow level is that of a compound nucleus, the lifetime of that level can be as long as 10^{-15} sec or 10^{-14} sec, as mentioned earlier in this section. Along with the level width, the level spacing D, or mean distance between levels, can be obtained from resonance measurements and is an important quantity in nuclear spectroscopy. The level width and level spacing are useful not only for characterizing compound nuclei, but for any excited nuclear states, and can be applied to both bound and virtual levels.

The study of resonance phenomena in nuclear reactions can now be seen to have great importance because it provides information about the width and spacing of the energy levels of the compound nucleus. The dependence of the width and spacing on the mass number and the excitation energy of the nucleus provides a test of theories of nuclear reactions and nuclear models. The partial widths of a level of the compound nucleus give the relative probabilities for different modes of disintegration, and these probabilities also yield information about nuclear structure.

16-3 Cross sections for nuclear reactions. To study nuclear reactions in detail, it is necessary to have a quantitative measure of the probability of a given nuclear reaction. This quantity must be one which can be measured experimentally and calculated in such a way that the theoretical and experimental values can be compared readily. The quantity that is most often used for this purpose is the cross section of a nucleus for a particular reaction, usually denoted by σ with an appropriate subscript. The term "cross section" has already been met in connection with the attenuation of a γ -ray beam, when it was used as a measure of the probability that a photon is removed from the beam; the cross section was regarded there as a form of absorption coefficient. In the discussion of nuclear re-

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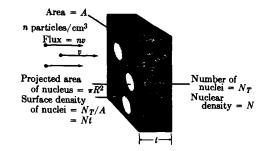


FIG. 16-2. Cross sections for nuclear reactions as geometrical areas $(Hughes^{(12)})$.

actions, a more detailed consideration of the idea of cross sections will prove useful. The concept of a nuclear cross section can be most easily visualized as the cross-sectional area, or target area, presented by a nucleus to an incident particle. If the nuclei are considered as spheres of radius R cm and the incident particles as point projectiles, then the target area, or cross section σ , of each nucleus is given by

$$\sigma = \pi R^2 \operatorname{cm}^2. \tag{16-9}$$

A particle which passes normally through a thin sheet of material of area A containing N_T nuclei has a probability $N_T \sigma/A$ of colliding with a nucleus provided that there is no "overlapping" of the nuclei, i.e., that $N_T \sigma/A$ is small (Fig. 16-2). The quantity N_T/A , which is the number of nuclei per square centimeter or the surface density of nuclei, is equal to Nt, where N is the number of nuclei per cubic centimeter and t is the thickness of the sheet. For an incident beam containing n particles per cubic centimeter, moving with velocity v, the number of particles passing through the sheet is nv per square centimeter per second, and the collision rate can be expressed as

collisions per square centimeter per second = $nv \cdot \frac{N_T \sigma}{A} = nv\sigma Nt;$

the collision cross section is then

$$\sigma = \frac{\text{collisions per square centimeter per second}}{nvNt} \cdot (16-10)$$

The quantity nv, which is the number of particles in the incident beam crossing one square centimeter of area each second, is called the *particle flux*. Equation (16-10) then shows that the cross section for collision is the number of collisions per unit volume per second for unit incident flux and unit nuclear density.

The above discussion can be applied to any nuclear reaction, and the term "collision" can be replaced by the appropriate term for any nuclear process. Thus, σ_s may denote the cross section for scattering of a given kind of particle, and may consist of two parts, σ_e , the cross section for elastic scattering, and σ_i , the cross section for inelastic scattering. It is possible to speak of a cross section σ_a for the absorption of a particular kind of particle, defined as the number of particles that are absorbed, or disappear, per cubic centimeter per second for unit incident flux and unit nuclear density. There are also cross sections for individual reactions. such as $\sigma(\alpha, p)$, $\sigma(\alpha, n)$, and $\sigma(p, \alpha)$. The number of particles removed from the beam is obtained by adding the numbers removed by all the processes which can take place; particles can be removed by being absorbed in nuclear reactions or by being scattered out of the beam. In an experiment, the number of events of a given kind (collisions, scatterings, absorptions, or other processes) is counted, and the quantities nv and N are measured; the cross section for the particular process is then given by a relationship of the type of Eq. (16-10). The collision cross section, which corresponds to the effect of all possible processes, is usually called the total cross section, σ_t and is the sum of the cross sections for the individual reactions. There is a direct analogy between σ_t and the total absorption coefficient for γ -rays, expressed in units of square centimeter per atom, which is the sum of a cross section for photoelectric absorption, a cross section for Compton scattering, and a cross section for pair formation. As in the case of γ -radiation, the total cross section can be found by measuring the transmission of a particle beam in a "good geometry" experiment.

The expression for the number of processes of a given kind, say the *i*th kind, contains the product $N\sigma_i$. This product represents the cross section, for the *i*th process, of all the atoms in a cubic centimeter of material. It is sometimes called the *macroscopic* cross section and denoted by

$$\sum_i = N\sigma_i;$$

it has the unit cm^{-1} .

A rough idea of the magnitude of cross sections for nuclear reactions can be obtained from Eq. (16-9) for the geometrical cross section of a nucleus. In the discussion of the theory of α -decay, it was found that the radii of α -emitting nuclei may be represented by the formula

$$R = 1.5A^{1/3} \times 10^{-13} \,\mathrm{cm}$$

where A is the atomic weight. If it is assumed that this formula may be applied to all nuclei, which will be shown to be a reasonably good approximation, then

$$\sigma = \pi R^2 = \pi (1.5)^2 A^{2/3} \times 10^{-26} \,\mathrm{cm}^2.$$

For a nucleus of intermediate mass, say A = 125,

$$\sigma = \pi (2.25)(25) \times 10^{-26} \,\mathrm{cm}^2 = 1.8 \times 10^{-24} \,\mathrm{cm}^2$$

and the geometrical cross section of a nucleus is of the order of 10^{-24} cm². Cross sections for nuclear reactions are often expressed in terms of the unit 10^{-24} cm², and this unit is called the *barn*, abbreviated as b.

Although it is convenient and simple to introduce the cross section as a target area, and to get a rough idea of its magnitude by calculating the geometrical cross section, this procedure must not be taken seriously. The experimental meaning of the cross section comes from its use as a measure of the number of nuclear events which occur under a given set of experimental conditions, as expressed by Eq. (16-10) and the subsequent discussion. Nuclear cross sections are found to have values ranging from small fractions of a barn to hundreds of thousands of barns, and these values often differ greatly from the geometrical cross section. A given nucleus can have widely different cross sections for different nuclear reactions and the values represent the relative probabilities of those reactions. Under certain special conditions scattering cross sections and geometrical cross sections can be related directly, and the measured values of the scattering cross sections are then used to determine the values of the nuclear radii. The elastic scattering of neutrons with energy greater than 10 Mey is a case in point, and will be discussed in Section 16-5E.

The results of an experimental study of a nuclear reaction can be expressed in terms of the number of processes which take place under the conditions of the experiment or in terms of the cross section for the reaction. The advantage of using the cross section lies in the fact that its value is independent of the flux of incident particles and the density of the material used for the target, and these quantities are not essential to the nuclear reaction itself. When the cross section for a particular reaction between a given particle and a given nuclide is known, it is possible to predict the number of reactions that will take place when a sample of the nuclide is exposed to a known flux of those particles for a given length of time. This kind of information is needed in many practical problems. as in the manufacture of artificial radionuclides, and cross sections are a valuable form of nuclear data. In many cases, the results of the study of a nuclear reaction are expressed directly in terms of the number of processes that take place, or of the number of outgoing particles. If the main interest is in the determination of the energies of the different groups of particles emitted, it is enough to measure the energies by measuring the ranges or by magnetic deflection, without knowing exactly how many processes have taken place. The relative intensities of different groups of product particles can also be measured by methods which are independent of the number of processes, as can the energies of resonances in nuclear reactions.

The concepts of cross section and level width can be applied to resonances in a quantitative way. The probability of the reaction X(x, y)Y may be denoted by the cross section $\sigma(x, y)$. According to the two-step view of nuclear reactions,

$$\sigma(x, y) = \sigma_C(x) \cdot (\text{relative probability of the emission of } y), \quad (16-11)$$

where $\sigma_C(x)$ is the cross section for the formation of the compound nucleus in a collision between the particle x and the target nucleus X. The relative probability of the emission of the particle (or γ -ray) y is just Γ_y/Γ , where Γ_y is the partial level width for y and Γ is the total level width. Then

$$\sigma(x, y) = \sigma_C(x) \frac{\Gamma_y}{\Gamma}$$
 (16-12)

In general, the values of the cross sections and level widths depend on the energy of the incident particle, and on the charge and mass of the target nucleus. One of the problems of nuclear theory is the calculation of $\mathcal{G}_C(x)$, Γ , Γ_y , and therefore of $\sigma(x, y)$. In the particularly important case of resonance processes, a theoretical formula for the cross section was derived by Breit and Wigner.^(9,10) The rigorous derivation of the Breit-Wigner formula is a difficult problem, but the physical meaning of the formula can be discussed qualitatively. In its simplest form, the Breit-Wigner formula gives the value of the cross section in the neighborhood of a single resonance level formed by an incident particle with zero angular momentum. Under these conditions, the formula is

$$\sigma(x, y) = \frac{\lambda^2}{4\pi} \frac{\Gamma_x \Gamma_y}{(E - E_0)^2 + (\Gamma/2)^2}, \qquad (16-13)$$

where λ is the de Broglie wavelength of the incident particle defined in Section 7-8, *E* is the energy, E_0 is the energy at the peak of the resonance, and the Γ 's are the widths already defined. Equation (16-13) may be regarded as containing three factors. The first is a measure of the probability of forming a compound nucleus and, according to wave mechanics, is proportional to λ^2 . The second factor,

$$\frac{1}{(E-E_0)^2+(\Gamma/2)^2}$$

is the mathematical expression for the resonance property and may be called the *resonance factor*. The denominator of this factor has its smallest value when $E = E_0$, and the cross section then has its greatest value. As the value of E departs from E_0 , the denominator increases in value,

and the resonance factor and cross section decrease. The third factor is the probability for definite types of disintegration of the compound nucleus and is expressed by the partial widths Γ_x and Γ_y .

The Breit-Wigner formula is an example of the application of wave mechanics to nuclear physics, and it is typical of such applications that the wavelength of the incident particle appears in the formula. The de Broglie wavelength of a particle is defined as

$$\lambda = \frac{h}{mv}, \qquad (16-14)$$

where h is Planck's constant and mv is the momentum of the particle. It is convenient to express λ in terms of the kinetic energy rather than the momentum and, since $E = \frac{1}{2}mv^2$,

$$\lambda = \frac{h}{\sqrt{2mE}}.$$
 (16–15)

An idea of the magnitudes of λ and σ may be obtained by considering the incident particle to be a neutron; when the numerical values of h and m are inserted, and E is expressed in electron volts, the result is

$$\lambda$$
 (cm) = $\frac{2.87 \times 10^{-9}}{\sqrt{E (\text{ev})}}$. (16-16)

A neutron with a kinetic energy of 10^4 ev has a wavelength of 2.87×10^{-11} cm, while a 1-Mev neutron has a wavelength of 2.87×10^{-12} cm. Since $\sigma(x, y)$ is proportional to λ^2 , its value should be of an order of magnitude similar to that obtained from the geometrical estimate in this section. Its actual value in any particular case will, of course, depend also on the other quantities in Eq. (16-13). It is found that the values of resonance cross sections vary over a wide range, from fractions of a barn to the order of 10^6 barns.

Values of $\sigma_0(x, y)$ (the cross section at the peak of the resonance), E_0 , and Γ can be obtained from experiments in which the particle transmission through the target is measured as a function of the energy of the incident particles in the neighborhood of the resonance energy. The values of σ_0 and E_0 can be deduced from the transmission curve. It can be shown from Eq. (16–13) that the "full width at half-maximum," that is, the energy interval between the two points at which the cross section is $\sigma_0/2$, is just the total width Γ . The total width can thus be determined directly from the actual observed width of the cross section resonance. When the total width is known, the values of the partial widths can be obtained by further experiments, or may sometimes be inferred with the aid of certain theoretical considerations.

Chapter 3

NUCLEAR FISSION

19-1 The discovery of nuclear fission. The discovery of nuclear fission was one of the results of attempts to make transuranium elements of atomic number greater than 92 by means of (n, γ) reactions followed by β -decay of the product nucleus. These attempts eventually succeeded, and some of the methods used and results obtained were discussed in Section 12-3. The interpretation of the early experiments was difficult, however, because of unexpected results which could be explained only in terms of nuclear fission.

To understand some of the difficulties that were met, it is necessary to consider briefly how radioactive elements are separated from inactive ones. A radioelement formed by a nuclear reaction is usually available only in a very small amount, possibly as small as 10^{-12} gm, and cannot be separated by means of ordinary chemical methods. But the separation can often be made with the aid of a *carrier* which is a stable substance with chemical properties similar to those of the radioelement. The element and the carrier usually belong to the same subgroup of the periodic system and can undergo similar chemical reactions. If an appreciable amount, perhaps 10 to 100 mg, of the carrier is added to a solution containing the radioelement, and if the carrier is then precipitated from the solution by the formation of an insoluble salt, the radioelement can then be separated by means of other chemical reactions.

In the separation of radium from other members of the uranium series, barium serves as a carrier. Radium and barium both belong to group IIA of the periodic system and form insoluble sulfates. When a solution of a barium salt is added to a solution containing a very small amount of radium and a sulfate is added, barium and radium sulfates are precipitated together. A neater method depends on the fact that barium and radium chlorides are precipitated together from concentrated solutions of hydrochloric acid. The precipitate can be dissolved in water, and the barium and radium can then be separated by repeated fractional crystalization from hydrochloric acid. In analogous ways, lanthanum acts as a carrier for actinium (Z = 89). Sometimes the carrier is a stable form of the radioelement, e.g., stable iodine can be used as a carrier for radioactive isotopes of iodine.

In the early experiments on the formation of transuranium elements, uranium was bombarded with neutrons and several different β -activities,

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distinguished by their half-lives, were detected. Carrier techniques were used to separate the elements responsible for the activities, but the number and properties of the new radioelements were such that they could not be fitted into a scheme consistent with the known properties of the heavy elements and the predicted properties of the transuranium elements. One difficulty in the analysis of the products of the bombardment of uranium with neutrons led to a remarkable conclusion. In addition to the elements which seemed to be real transuranium elements, there were four which were supposed to be β -radioactive isotopes of radium because they were precipitated with barium when the latter was used as the carrier. The decay products of these nuclides seemed to be isotopes of actinium because they were precipitated with lanthanum, the carrier for actinium. Although these results appeared to be consistent, since actinium follows radium in the periodic system, they raised two serious questions. First, the production of an isotope of radium by the neutron bombardment of uranium would require an $(n, 2\alpha)$ reaction, but this reaction is a very unlikely one (cf. Section 16-5), especially at low neutron energies. Second. further chemical experiments showed that the "radium" activities could not be separated from the barium carrier, and the daughter activities could not be separated from the lanthanum carrier. In 1939, Hahn and Strass $mann^{(1,2)}$ performed a beautiful and thorough set of experiments which proved beyond a doubt that the "radium" isotopes are really isotopes of barium and the "actinium" isotopes are isotopes of lanthanum. Furthermore, they showed that one of the barium isotopes resulting from the neutron bombardment of uranium could be identified, because of its halflife of 86 min, with the previously known nuclide Ba¹³⁹, which has the same half-life. Similarly, one of the lanthanum isotopes from the neutron bombardment of uranium was identified with the known nuclide La¹⁴⁰. which has a half-life of 40 hr.

The production of the nuclides ${}_{57}La^{140}$ and ${}_{56}Ba^{139}$ from uranium, which has the atomic number 92 and an atomic weight of nearly 240, required a hitherto unknown kind of nuclear reaction in which the uranium nucleus is split into fragments which are themselves nuclei of intermediate atomic weight. If such a process really occurs, it should also be possible to find nuclei with masses between 90 and 100 and atomic numbers of about 35. Hahn and Strassmann were able to find an active isotope of strontium (Z = 38) and one of yttrium (Z = 39) which met these requirements, as well as isotopes of krypton (Z = 36) and xenon (Z = 54). It was clear from the chemical evidence that uranium nuclei, when bombarded with neutrons, can indeed split into two nuclei of intermediate atomic weight. It was then predicted, from the systematics of stable nuclides and from the semiempirical binding-energy formula, that the product nuclei would have very great energies and would produce large numbers of ion pairs in

NUCLEAR FISSION

passing through a gas. When a thin layer of uranium was put in a suitable ionization chamber connected to an amplifier and irradiated with neutrons, great bursts of ionization were observed^(3, 4) corresponding to energies up to 100 Mev. These pulses of ionization are extremely large compared with those of single α -particles and are easy to recognize. Further chemical work then showed that besides the reaction products mentioned above, other elements of medium mass number were formed, including bromine, molybdenum, rubidium, antimony, tellurium, iodine, and cesium. There was, therefore, ample chemical and physical evidence for the splitting of the uranium nucleus, and this process was called *fission*.⁽⁵⁾

It is now known that fission can be produced in various nuclides under different conditions, and some of the results will be mentioned. When small samples of the separated isotopes of uranium, prepared in a mass spectrograph, were bombarded, it was found⁽⁶⁾ that slow neutrons cause fission of U²³⁵ but not of U²³⁸; fast neutrons, with energies greater than one Mev, cause fission of both U²³⁵ and U²³⁸. Thorium and Pa²³¹ undergo fission, but only when bombarded with fast neutrons. Fission can also be produced in uranium and thorium by high-energy α -particles, protons, deuterons, and γ -rays. The nuclei Pu²³⁹ and U²³³, formed by (n, γ) reactions on U²³⁸ and Th²³², respectively, followed by β -decay of the products, undergo fission when bombarded with slow or fast neutrons, as do other artificial heavy nuclides. Finally, some heavy nuclei have been found to undergo spontaneous fission; in this process, the nucleus divides in the ground state without bombardment by particles from outside.

In addition to the two large fission fragments, neutrons and γ -rays are emitted. Division into three fragments of comparable size (ternary fission) has been observed but is a very rare event, occurring about five times per million binary fissions. Long-range α -particles are sometimes emitted, about once in every 400 fissions. The emission of light nuclei, with masses greater than 4 and probably less than 12, is a relatively common event, occurring once in about every 80 fissions.

19-2 Fission cross sections and thresholds. The probability of fission, as compared with that of other reactions, is a matter of theoretical interest and practical importance. The U^{235} nucleus may capture a thermal neutron to form the compound nucleus $[U^{236}]$, or the neutron may be scattered. The compound nucleus may either undergo fission, or it may emit γ -rays and decay to the ground state of U^{236} , which emits a 4.5-Mev α -particle and has a half-life of 2.4×10^7 years. The thermal cross sections⁽⁷⁾ for the different reactions for U^{235} , U^{238} , natural uranium, and Pu^{239} are given in Table 19-1. The total cross section of U^{235} for 2200 m/sec neutrons is 698 b, the total absorption cross section is 683 b, and the fission cross section is 577 b. The ratio of the radiative capture cross section to the

TABLE 19-1

	U ²³³	U ²³⁵	Natural Uranium	Pu ²³⁹
σ_{abs} σ_{f} σ_{r} σ_{s} ν (average number of	$578 \pm 4 \\ 525 \pm 4 \\ 53 \pm 2 \\ 2.51 \pm 0.02$	$\begin{array}{c} 683 \pm 3 \\ 577 \pm 5 \\ 101 \pm 5 \\ 15 \pm 2 \\ 2.44 \pm 0.02 \end{array}$	$7.68 \pm 0.07 4.18 \pm 0.06 3.50 8.3 \pm 0.2$	$ \begin{array}{r} 1028 \pm 8 \\ 742 \pm 4 \\ 286 \pm 4 \\ 9.6 \pm 0.5 \\ 2.89 \pm 0.03 \end{array} $
neutrons per fission) $\alpha = \sigma_{\tau}/\sigma_{f}$ $\eta = \frac{\nu}{1 + \alpha}$	0.101 ± 0.004 2.28 ± 0.02	0.18 ± 0.01 2.07 ± 0.01	1.34 ± 0.02	0.39 ± 0.03 2.08 ± 0.02

PROPERTIES OF FISSIONABLE MATERIALS (7,8)*

*For 2200 m/sec neutrons.

fission cross section is 0.18, so that the probability of radiative capture is about 18% that of fission. The natural abundance of U^{235} is only 0.72%, with the result that the fission and radiative capture cross sections of natural uranium are much smaller than those of the separated isotope U^{235} . In Pu²³⁹ the ratio of the capture cross section to that for fission is 0.39; although the fission cross section is greater than in U^{235} , the ratio of capture to fission is also greater.

Some of the artificially produced radioactive nuclides undergo fission with thermal neutrons, as do some of the shorter-lived naturally occurring radionuclides. Values of the fission cross section for 2200 m/sec neutrons are listed in Table 19–2, together with values of activation cross sections and total absorption cross sections where these are available.⁽⁷⁾ It is evident that many heavy nuclides are fissionable, and that the probability of fission varies over a wide range of values. Only U^{235} , U^{233} and Pu^{239} have high cross sections as well as long half-lives, and either occur naturally (U^{235}) or can be produced in significant amounts in practical lengths of time (Pu^{239} and U^{233}). Hence, only these three fissionable materials are important in the large-scale applications of nuclear fission.

The fission cross section varies with energy in a complicated way, as is shown in Fig. 19-1 for U^{235} . In the thermal region σ_f varies approximately as 1/v; starting at 0.28 ev, there are many closely spaced resonances, with at least 20 resolved resonances below 20 ev. At high energies, the fission cross section is relatively small, only about one barn in the neighborhood of 1 Mev; the fission cross sections of U^{233} and U^{235} in the

TABLE 19-2

THERMAL.	CROSS	SECTIONS	OF SOME	HEAVY	NUCLIDES ⁽⁷⁾ *

Nuclide	Half-life	Fission cross section, σ_f , b	Total absorption cross section, σ_a , b	Activation cross section, σ_{act} , b
90Th ²²⁷	18.2d	1500 ± 1000		
90Th ²²⁹	7.34×10^{3} y	45 ± 11		
91Pa ²³⁰	17.3d	1500 ± 250		
91Pa ²³¹	3.4×10^4 y	$(10 \pm 5) \times 10^{-3}$		200 ± 15
91Pa ²³²	1.3d	700 ± 100		760 ± 100
$_{92}\mathrm{U}^{230}$	20.8d	25 ± 10		
92U ²³¹	4.2d	400 ± 300		
$92U^{232}$	74y	80 ± 20		300 ± 200
$92U^{233}$	$1.62 imes 10^5$ y	525 ± 4	578 ± 4	53 ± 2
92U ²³⁴	$2.52 imes 10^5$ y	< 0.65	105 ± 4	90 ± 30
$92U^{235}$	7.1×10^{8} y	582 ± 4	683 ± 3	101 ± 5
93Np ²³⁴	4.4d	900 ± 300		
93Np ²³⁶	22h	2800 ± 800		
93Np ²³⁷	$2.2 imes 10^{6}$ y	$(19 \pm 3) \times 10^{-3}$	170 ± 5	169 ± 6
93Np ²³⁸	2.1d	1600 ± 100		
93Np ²³⁹	2.3d	<1	:	35 ± 10
94Pu ²³⁸	86.4y	16.8 ± 0.3		400 ± 10
94Pu ²³⁹	2.44×10^4 y	742 ± 4	1028 ± 8	286 ± 4
94Pu ²⁴⁰	$6.6 imes 10^{3} ext{y}$	0.030 ± 0.045	286 ± 7	250 ± 40
94Pu ²⁴¹	13y	1010 ± 13	1400 ± 80	400 ± 50
94Pu ²⁴²	$3.75 imes 10^5 ext{y}$	< 0.2	30 ± 2	19 ± 1
95Am ²⁴¹	458y	3.2 ± 0.2	630 ± 35	750 ± 80
95Am ²⁴²	100y	6400 ± 500		

*For 2200 m/sec neutrons.

neighborhood of 1 Mev are shown in Fig. 19–2a. Some heavy nuclides, for example, U^{234} , U^{236} , and U^{238} , do not undergo fission with slow neutrons, but only with fast neutrons. Fission is a threshold reaction in these nuclides, and the fission cross section varies with energy in much the same way that the cross sections for other threshold reactions do. The fast fission thresholds of U^{234} , U^{236} , and U^{238} , and the variation of the fission cross section with energy, above threshold, have been determined by bombarding the nuclides with neutrons from the reaction $H^3(p, n)He^3$. The results are shown in Fig. 19–2b.

Threshold energies and cross sections have been measured for fission processes induced by charged particles.⁽¹⁰⁾ The threshold for fission by deuterons is close to 8 Mev for Th²³², U²³⁵, and U²³⁸; that for fission by

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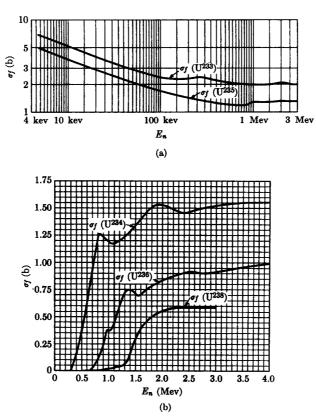


FIG. 19–2. The fission cross sections of several uranium isotopes in the Mev region.^(7,9) (a) U^{233} , U^{235} (b) U^{234} , U^{236} , U^{238} .

 α -particles is in the neighborhood of 21 Mev for these nuclides. In the range of energies explored so far, the value of the cross section increases with energy above the threshold and then either levels off or passes through a maximum. In the case of U²³⁵ bombarded with deuterons, the cross section rises to a value of one barn at about 20 Mev and then increases slowly to a value between 1.5 and 2 b at deuteron energies between 120 and 200 Mev. The cross section for the fission of U²³⁵ by α -particles rises to a value of 1.5 b at about 50 Mev and remains at about this value up to energies of 400 Mev.

Gamma-rays from nuclear reactions and high-energy x-rays from a betatron can cause fission, and some of the measured thresholds⁽¹¹⁾ are

TABLE 19–3

THRESHOLD ENERGIES FOR PHOTOFISSION

Nuclide	Photofission threshold, Mev		
92U ²³⁸ 92U ²³⁵ 92U ²³³ 94Pu ²³⁹ 90Th ²³²	$\begin{array}{c} 5.08 \ \pm \ 0.15 \\ 5.31 \ \pm \ 0.25 \\ 5.18 \ \pm \ 0.27 \\ 5.31 \ \pm \ 0.27 \\ 5.40 \ \pm \ 0.22 \end{array}$		

shown in Table 19-3. The cross sections for these reactions are generally smaller than those for neutron-induced fission. When U^{238} is bombarded by monoenergetic 6.3-Mev γ -rays from the $F^{19}(p, \gamma)Ne^{20}$ reaction, the cross section is about 3 mb; for the 17.5-Mev γ -rays from the $Li^7(p, \gamma)Be^8$ reaction, the cross section is about 30 mb.

Many heavy nuclides undergo spontaneous fission,⁽¹²⁾ and this process is an alternative, less probable, method of nuclear disintegration than α -particle emission. Nuclei which undergo fission with slow neutrons have smaller spontaneous fission rates than their isotopes which undergo fission only with fast neutrons. For example, U²³⁵ has a half-life for spontaneous fission of about 1.8×10^{17} years, corresponding to a rate of about one fission per gram per hour while U²³⁸ has a fission half-life of 8.0×10^{15} years, or a rate of 25 fissions/gm/hr. Similarly, Pu²³⁹ has a spontaneous fission rate of 36 fissions/gm/hr, or a fission half-life of 5.5×10^{15} years, while Pu²⁴⁰ has a rate of 1.6×10^6 fissions/gm/hr and a half-life of 1.2×10^{11} years.

Finally, fission can be produced in bismuth, lead, thallium, mercury, gold, and platinum by neutrons with very high energies, e.g., 40 Mev.

19-3 The fission products. It was indicated in Section 19-1 that a number of nuclides of intermediate charge and mass are formed when a uranium nucleus undergoes fission. The study of the nuclei formed in fission was evidently a promising source of information about the mechanism of the fission process and offered the possibility that new, hitherto unknown, nuclides might be discovered. The latter possibility became apparent when the neutron-to-proton ratios of the uranium isotopes were compared with those of some of the fission products. The compound nucleus $[9_2U^{236}]$ has 144 neutrons and 92 protons, and the value of the ratio is 144/92 = 1.57. The values of the ratio for the stable isotopes of some typical fission products (krypton, iodine, xenon, and cesium) vary

from 1.17 to 1.52, and are appreciably lower than that for $[U^{236}]$. When that excited nucleus splits into two smaller nuclei, the neutron-to-proton ratio for at least one of them must be greater than the value compatible with stability. Such an unstable nucleus might be expected to approach stability by electron emission or, if the excitation energy is high enough, by ejection of one or more neutrons; it has been found experimentally that both of these processes occur.

The investigation of the products of the fission of U^{235} has shown that the range of mass numbers is from 72, probably an isotope of zinc with atomic number 30, to 158, thought to be an isotope of europium with atomic number 63. About 97% of the U^{235} nuclei undergoing fission yield products which fall into two groups, a "light" group with mass numbers from 85 to 104, and a "heavy" group with mass numbers from 130 to 149. The most probable type of fission, which occurs in about 7% of the total, gives products with mass numbers 95 and 139. There are 87 possible mass numbers between 72 and 158, which may represent the total number of different nuclides formed as direct fission fragments. If this were the case, the uranium nucleus should be capable of splitting in over 40 different ways. More than 60 primary products have actually been detected, so that there are at least 30 different modes of fission, a different pair of nuclei being formed in each mode.

The fission fragments have too many neutrons for stability and most of them decay by electron emission. Each fragment starts a short radioactive series, involving the successive emission of electrons. These series are called *fission decay chains*, and each chain has three members, on the average, although longer and shorter chains occur frequently. The problem of determining the masses and atomic numbers of the fission products and of identifying the members of the many decay chains is an extremely difficult one. Nevertheless, as the result of careful and persistent work, more than 60 chains have been established, and about 200 different radionuclides have been assigned to them.^(13,14) An example of a long chain is

$$_{54} Xe^{140} \xrightarrow{\beta^{-}}_{16s} {}_{55} Cs^{140} \xrightarrow{\beta^{-}}_{66s} {}_{56} Ba^{140} \xrightarrow{\beta^{-}}_{12.8d}$$

 $\longrightarrow {}_{57} La^{140} \xrightarrow{\beta^{-}}_{40h} {}_{58} Ce^{140} (stable).$

This chain is especially interesting because it contains two of the nuclides, Ba^{140} and La^{140} , whose appearance led to the discovery of fission. The short chain

$$_{60}$$
Nd¹⁴⁷ $\xrightarrow{\beta^{-}}_{11d}$ $_{61}$ Pm¹⁴⁷ $\xrightarrow{\beta^{-}}_{4y}$ $_{62}$ Sm¹⁴⁷ (~10¹¹ y)

is important because one of its members is an isotope of the element with

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atomic number 61. This element had not been clearly isolated before the discovery of fission, and has now been named *promethium* (symbol, Pm). Radioactive isotopes of the element with atomic number 43, which has not been found to occur in nature, also have been identified as fission products. This element is now called *technetium* (symbol, Tc), and its longest-lived isotope occurs in the chain.

$$_{42}Mo^{99} \xrightarrow{\rho^-}_{66h} _{43}Tc^{99} \xrightarrow{\rho^-}_{2.2 \times 10^5 y} _{44}Ru^{99}$$
(stable).

19-4 The mass and energy distributions of the fission products. The mass distribution of the fission products is shown most conveniently in the form of a *fission yield curve*, in which the percentage yields of the different products are plotted against mass number. The yield of any given mass can be found by measuring the abundance of a long-lived nuclide near the end of a chain, or that of the stable end product. Yield curves^(15,16) for the fission of U²³⁵ by thermal neutrons and by 14-Mev neutrons are shown in Fig. 19-3a; curves for thermal fission of U²³³ and Pu²³⁹ are shown in Fig. 19-3b. The fission yield for a particular nuclide is the probability (expressed as a percentage) of forming that nuclide or the chain of which it is a member; it may also be regarded as the percentage of fission, the total yield adds up to 200%. The yields vary from about $10^{-5}\%$ to about 7%,

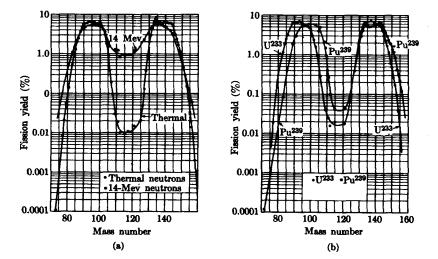


FIG. 19-3. Fission yields from U²³⁵, U²³³, and Pu²³⁹: (a) thermal and fast fission of U²³⁵; (b) thermal fission of U²³³ and Pu^{239.(15)}

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and the range is so large that a logarithmic scale is used for the ordinate of the yield curve. Each curve shows two peaks, corresponding to the light and heavy groups of products. In the case of U^{235} the maxima lie near mass numbers 95 and 135; fission induced by slow neutrons is a highly asymmetric process, and division into two equal fragments occurs in only about 0.01% of the fissions. Small "fine-structure" peaks are evident in some of the yield curves; for U²³⁵, these peaks are at masses 100 and 134. This fine structure is, for the most part, a shell effect, and can be accounted for by a detailed analysis of the neutron binding energies in nuclei with neutron numbers close to those corresponding to closed shells. i.e., 50 and 82 neutrons. The U²³⁵ curves illustrate the effect on the mass distribution of increasing the neutron energy above thermal. The greatest change is the increase in the probability of symmetric fission; for fission by 14-Mev neutrons, the increase is about 100-fold. The other changes are a small drop in the peak yields and a moderate increase in the most asymmetric modes of fission, i.e., a rise in the wings of the yield versus mass curve.

The mass distribution of the fission fragments can also be obtained from the distribution of their kinetic energies, which can be determined by measuring the ionization produced in an appropriate ionization chamber. In one type of chamber, the fissile material is placed on one of the electrodes and the ions which result when a fission fragment enters the region between the electrodes are collected. Since the fission fragments occur in pairs, an experiment of this kind measures the energy of one fragment only. In another type of chamber, a very thin foil is made the common cathode of two back-to-back ionization chambers. The two fragments resulting from neutron bombardment travel in opposite directions into the two chambers and the ionizations they cause are measured simultaneously. The nucleus undergoing fission can be considered to be initially at rest; and, if the neutrons emitted are neglected, the law of conservation of momentum gives

$$M_1 V_1 = M_2 V_2, (19-1)$$

where the subscripts refer to the two fission fragments. The energies of the fragments are then in the ratio

$$\frac{E_1}{E_2} = \frac{\frac{1}{2}M_1V_1^2}{\frac{1}{2}M_2V_2^2} = \frac{M_2}{M_1},$$
(19-2)

and the masses are inversely proportional to the kinetic energies; when the energy distribution has been measured, the mass distribution is obtained from Eq. (19-2).

The energy distribution of the fission products has been measured for the fission of U^{233} , U^{235} , and Pu^{239} by thermal neutrons, (17, 18) and for

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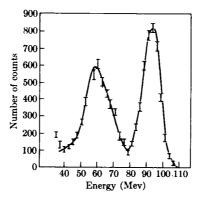


FIG. 19-4. Energy distribution of the fragments from fission of U^{235} by thermal neutrons (Brunton and Hanna⁽¹⁷⁾).

the fission of U^{235} , U^{238} , Th^{232} , and Pu^{239} by fast neutrons.^(19,20) The energy distribution in the case of the thermal fission of U²³⁵ is shown in Fig. 19-4; this energy spectrum is typical for fission induced by slow neutrons, and the curves for U²³³ and Pu²³⁹ are similar to the curve shown. The peaks near 60 Mev and 95 Mev show the asymmetry of the fission process. The best ionization chamber measurements⁽¹⁷⁾ give 154.7 Mey for the average total kinetic energy of the fragments from U^{235} . The energy distribution for fission induced by fast neutrons (2.5 and 14 Mev) show the same two peaks, and the maxima differ only slightly from the values for fission induced by slow neutrons. The most important difference in the shapes of the spectra is an increase in the height of the curve in the region between the two peaks when the energy of the bombarding neutrons is in the Mev range. This rise is interpreted as indicating increased probability of symmetrical fission when the neutron energy is high. In the case of U^{235} , symmetrical fission is about 100 times more probable for 14-Mev neutrons than for thermal neutrons. When the energy of the bombarding neutrons is raised to 45 Mey, there are still two peaks, but the dip between them is small, and the probability of symmetrical fission is still greater than at the lower energies. With 90-Mev neutrons, only one peak is observed, corresponding to division into two equal fragments; at these very high energies symmetrical fission is the most probable mode.⁽²¹⁾

The velocity distribution of the fission fragments has been studied directly by means of a time-of-flight method.⁽²²⁾ A thin foil with a film of U^{233} , U^{235} , or Pu^{239} is irradiated by thermal neutrons, and pairs of fragments are detected by scintillation detectors. One fragment travels only about 1 cm before striking a detector, while the other travels about

350 cm along an evacuated tube. The pulses from the two detectors are shown on the screen of a cathode-ray tube and photographed; the velocity of the fragment can be obtained from the distance between the two peaks, and the kinetic energy is then calculated from the velocity. Results obtained with this method show that the energies are greater than those given by the ionization measurements, and the average total kinetic energy of the fission fragments from U^{235} is increased by 12.4 Mev, from 154.7 Mev to 167.1 Mev. This value agrees with the result, 167.1 \pm 1.6 Mev, of a calorimetric measurement.⁽²³⁾

The energy distribution of the fragments from spontaneous fission has also been determined⁽²⁴⁾ and is similar to that found in neutron-induced fission.

The determination of the charge distribution in fission is more difficult than that of the energy distribution because in an ionization chamber or cloud chamber there is no obvious way of determining the nuclear charge at the instant of fission. Radiochemical methods based on attempts to measure the primary fission yield have been used;^(16,25) these methods are indirect, and the problem has not yet been solved.⁽²⁶⁾

19-5 Neutron emission in fission. The comparison of the value of the neutron-to-proton ratio of the compound nucleus [U²³⁶] with the values for some of the fission products, discussed in Section 19-3, indicated also that neutrons might be emitted in fission. The fact that neutrons are emitted was shown in a simple experiment shortly after the discovery of fission.⁽²⁶⁾ A neutron source was placed at the center of a large vessel, with detectors at various distances from it to determine the neutron density in the vessel. The vessel was filled first with a uranyl sulfate solution, and then with an ammonium nitrate solution for comparison. The average neutron density was found to be greater when uranium was present, showing in a rough way that more neutrons were formed, when fission occurred, than were used up. The average value of the number of neutrons released in fission, usually denoted by ν , has been measured for various fissionable materials, and some of the results^(27,28) are listed in Table 19-4. The average number of neutrons released is always greater than two and increases with the energy of the neutrons that induce fission. The number of neutrons released in any one fission process must, of course, be an integer but, since the fissionable nucleus can divide in at least 30 different ways, the average value ν of the number of neutrons does not have to be an integer.

In addition to ν , there is another property of fissionable materials which has practical importance, the average number of neutrons emitted per neutron absorbed by a fissionable nuclide. It is evident from the cross section values of Table 19-1 that all of the neutrons absorbed by a fis-

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TABLE 19-4

Neutron energy, Mev	U ²³³ + n	U ²³⁵ + n	U ²³⁸ + n	Pu ²³⁹ + n
Thermal	2.51 ± 0.02	2.44 ± 0.02		2.89 ± 0.03
0.08	2.58 ± 0.06	2.47 ± 0.03		3.05 ± 0.08
1.3	2.69 ± 0.05	2.61 ± 0.09		3.08 ± 0.05
1.5		2.57 ± 0.12	2.65 ± 0.09	
1.8	2.75 ± 0.06	2.72 ± 0.06		3.28 ± 0.06
2.0		2.80 ± 0.15		
4.0	3.06 ± 0.12	3.01 ± 0.12	3.11 ± 0.10	3.43 ± 0.11
14.1	3.86 ± 0.28	4.52 ± 0.32	4.13 ± 0.25	4.85 ± 0.50

THE AVERAGE NUMBER OF FISSION NEUTRONS AS A FUNCTION OF THE ENERGY OF THE NEUTRONS INDUCING FISSION⁽²⁷⁾

sionable material do not induce fission; some absorptions result in the emission of γ -rays, i.e., radiative capture competes with fission. With U^{235} , for example, the reaction $U^{235}(n, \gamma)U^{236}$ also occurs, and U^{236} is an α -emitter with a half-life of 2.4×10^7 years. The ratio of the radiative capture cross section to the fission cross section is usually denoted by α ,

$$\alpha = \sigma_r / \sigma_f, \qquad (19-3)$$

and the number of fission neutrons released per neutron absorbed in a fissionable nuclide is given by

$$\eta = \nu/(1+\alpha) \cdot \tag{19-4}$$

Values of α and η for thermal neutrons are listed in Table 19-1 for the important fissionable materials.

The average number of neutrons emitted per spontaneous fission has been determined for various heavy nuclides.⁽¹²⁾ The rate (number of fissions per gram per second) can be measured⁽²⁹⁾ as well as the total number of neutrons.⁽³⁰⁾ For U²³⁸, the rate is (6.90 \pm 0.24) \times 10⁻³ fissions/gm/sec, or about 25 fissions/gm/hr, and $\nu = 2.4 \pm 0.2$, close to the value for fission induced by thermal neutrons; for Th²³², $\nu = 2.6 \pm$ 0.3, and for Cf²⁵², $\nu = 3.53 \pm 0.15$.

The neutrons emitted as a result of the fission process can be divided into two classes, prompt neutrons and delayed neutrons. The prompt neutrons, which make up about 99% of the total fission neutrons, are emitted within an extremely short interval of time, possibly as low as 10^{-14} sec, of the fission process. It is thought that the compound nucleus $[U^{236}]$ first splits into two fragments, each of which has too many neutrons for stability, and has also the excess energy (6 Mev or more) needed to expel a neutron. The excited, unstable nucleus consequently ejects

NEUTRON EMISSION IN FISSION

TABLE 19-5

Half-life,	Energy, Mey	Yield, % of total neutrons e Thermal and fast fission			emitted in fission, Fast fission		
sec	Mev	U ²³⁵	U ²³³	Pu ²³⁹	U ²³⁸	Th ²³²	
55	0.25	0.021	0.022	0.007	0.020	0.075	
22	0.46	0.140	0.078	0.063	0.215	0.330	
5.6	0.41	0.125	0.066	0.046	0.254	0.341	
2.1	0.45	0.253	0.072	0.068	0.609	0.981	
0.6	0.42	0.074	0.013	0.018	0.353	0.378	
0.2		0.027	0.009	0.009	0.118	0.095	
Total	yield	0.64	0.26	0.21	1.57	2.20	

PROPERTIES OF DELAYED NEUTRONS⁽³³⁾

one or more neutrons within a very short time after its formation; prompt γ -rays are apparently emitted at the same time. These ideas are consistent with the results of experiments on the angular correlation between the direction of the neutrons and that of the fragments,⁽³¹⁾ the neutrons being emitted preferentially in the same direction as the fragments.

The delayed neutrons, which constitute about 0.64% of the total neutrons from the fission of U^{235} , are emitted with gradually decreasing intensity for several minutes after the actual fission process. Six well-defined groups of delayed neutrons have been observed by studying the rate of decay of the neutron intensity.^(32,33) The rate of decay of each group is exponential, just as for other forms of radioactive change, and a specific half-life can be assigned to each group, as well as a mean life and decay constant. From the intensity, the fraction β_i , which the group constitutes of the total (prompt and delayed) fission neutrons can be determined. The properties of the prominent groups of delayed neutrons are listed in Table 19–5. Three additional low-intensity delayed neutron groups from uranium have been reported, ⁽³⁴⁾ with half-lives of 3, 12, and 125 min and yields, per fission, of 5.8×10^{-8} , 5.6×10^{-10} , and 2.9×10^{-10} .

The same groups of delayed neutrons are found in the fission of different heavy nuclides by slow or by fast neutrons, but the yields vary from nuclide to nuclide. Of the three important fissionable materials, U^{235} has the greatest yield, 0.64%, about three times that of Pu^{239} . Although the yield of delayed neutrons is less than one percent of the total number of neutrons emitted, the delayed neutrons have a strong influence on the time-dependent behavior of a chain-reacting system based on fission and play an important part in the control of the system.

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NUCLEAR FISSION

The mechanism for the emission of delayed neutrons is understood. Since some of the fission products are rich in neutrons and are very unstable with respect to β -emission, a product with Z protons and N neutrons may have a β -decay energy greater than the binding energy of the last neutron in the daughter product with Z + 1 protons and N - 1 neutrons. In the β -decay, the product nucleus can be left either in the ground state or in one of the many excited states. If the excitation energy of one of the excited states of the Z + 1, N - 1 nucleus is greater than the binding energy of the last neutron, de-excitation may occur by the emission of a neutron, leaving a nucleus with Z + 1 protons and N - 2 neutrons. The neutron emission will be delayed and will appear to have the half-life of the β -decaying nuclide (Z, N), i.e., that of the delayed neutron precursor. Several of the delayed neutron groups have been related to the decay of bromine and iodine fission products.⁽³⁵⁻³⁹⁾ Thus, Br⁸⁷ is the precursor of the 55-sec delayed neutron emitter. Iodine-137, with a half-life of 24 sec, is associated with the 22-sec group. This group may consist of two or more precursors with similar half-lives, and may also contain Br⁸⁸ which has a half-life of 16.3 sec. The 5.6-sec group includes I^{138} ($T_{1/2}$ = 6.3 sec) and possibly $Br^{89}(T_{1/2} = 4.4 \text{ sec})$. The decay scheme of Br^{87} is shown in Fig. 19–5. Some 70% of the β -decays of Br⁸⁷ go to the 5.4 Mev excited level of Kr⁸⁷; this level lies about 0.3 Mev above the level corresponding to the binding energy of the last (51st) neutron in Kr⁸⁷.

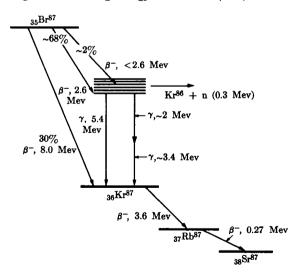


FIG. 19-5. Decay scheme of Br^{87} and its products, showing delayed neutron emission.

About 3% of the Kr^{87} nuclei in the 5.4 Mev level emit a neutron in the transition $\mathrm{Kr}^{87} \to \mathrm{Kr}^{86} + n + 0.3$ Mev, while the rest decay by γ -emission to the ground state of Kr^{87} . The *delayed neutrons* are delayed entirely by the 55-sec half-life of their β -ray parent Br^{87} . It should be noticed that Kr^{87} has 51 neutrons and the emission of the delayed neutron leaves a closed shell of 50 neutrons. Similarly ${}_{53}\mathrm{I}^{137}$ decays by β -emission to ${}_{54}\mathrm{Xe}^{137}$ which can emit a neutron, leaving ${}_{54}\mathrm{Xe}^{136}$, which has a closed shell of 82 neutrons; Br^{89} and I^{139} yield, on β -decay followed by n-emission, nuclei with 50 + 2 and 82 + 2 neutrons, respectively, which are again especially stable neutron configurations.

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19-6 The energy distribution of the neutrons emitted in fission. The energies of the prompt neutrons emitted in fission vary from values less than 0.05 Mev to more than 17 Mev. The determination of neutron energies over so wide a range is not an easy problem, and several methods have been used in order to cover the range. The neutrons from the thermal fission of U^{235} have been studied in great detail, and this case provides a good example of the methods and results.⁽⁴⁰⁾ In the energy range 0.05 to 0.7 Mev, a cloud chamber was used containing hydrogen gas and water vapor.⁽⁴¹⁾ A thin foil of U^{235} was bombarded with a beam of thermal neutrons from the thermal column of a nuclear reactor. The fission neutrons were allowed to enter the cloud chamber, where they collided with protons, and the range of the recoil protons was determined by measuring the track lengths. The energy distribution of the recoil protons of the neutrons was obtained from that of the recoil protons. The results

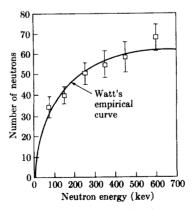


FIG. 19-6. Energies of fission neutrons: the low-energy region (Bonner et al.⁽⁴¹⁾).

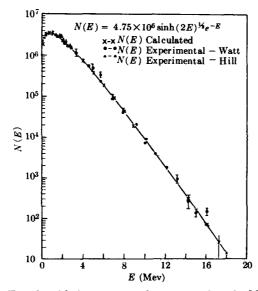


FIG. 19–7. Energies of fission neutrons: the spectrum from 0.4 Mev to 17 Mev (Watt⁽⁴³⁾).

are shown in Fig. 19-6, where the relative number of neutrons in a given energy interval is plotted against the energy and compared with an empirical formula which will be given below.

The neutron energy spectrum between 0.4 Mev and 17 Mev was studied by measuring, with coincidence counters, the range distribution of recoil protons from hydrogenous materials.^(42,43) The neutron intensity shows a broad maximum near 0.75 Mev, and decreases nearly exponentially at energies greater than 2 Mev. The energy distribution between 0.075 Mev and 17 Mev can be described by the empirical formula

$$N(E) = e^{-E} \sinh(2E)^{1/2}, \qquad (19-5)$$

where N(E) represents the relative number of neutrons per unit energy range as a function of the neutron energy. The experimental results between 0.4 Mev and 17 Mev are shown in Fig. 19-7, and agree with the empirical formula. The solid curve of Fig. 19-6 represents the same empirical formula, and the agreement is good also for the low-energy neutrons. The average value of the neutron energy is 2.0 \pm 0.1 Mev.

The fission neutron spectra of U^{235} and Pu^{239} , in the range from 0.5 to 8 Mev, have been studied with nuclear emulsion plates, ^(44,45) and the

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results agree with those just discussed. The spectrum for the neutrons from Pu^{239} shows a maximum in the 0.6- to 0.8-Mev region and the empirical formula of Eq. (19-5) fits the experimental results; the average energy is again 2.0 Mev.

19-7 The energy release in fission. One of the most striking properties of the fission process is the magnitude of the energy released per fission, which is about 200 Mev as compared with several Mev for other nuclear reactions. An estimate of the amount of energy released per fission can be made by considering the binding energy curve, Fig. 9-11. The value of the average binding energy per particle has a broad maximum of about 8.4 Mev in the range of mass numbers from 80 to 150, and it has been shown that nearly all of the fission products have mass numbers in this range. The average binding energy per particle is about 7.5 Mev in the neighborhood of uranium. Hence, the average binding energy per particle is about 0.9 Mev greater in the fission products than in the compound nucleus $[U^{236}]$, and the excess, 0.9 Mev, is liberated in the fission process. The total amount of energy released per fission should be roughly equal to the product of the number of particles (236) multiplied by the excess binding energy per particle (0.9 Mev), or approximately 200 Mev.

The total energy release per fission can also be calculated from the nuclear masses of $[U^{236}]$ and a typical pair of fission products. It has been shown that the fission products with the greatest yields have mass numbers near A = 95 and A = 139. If $_{42}Mo^{95}$ and $_{57}La^{139}$ are taken as a pair of stable products at the ends of the chains for their respective masses, their combined mass is 138.955 amu + 94.946 amu = 233.900 amu, as given by the semiempirical mass formula. The corresponding mass number is 234, and 2 neutrons are apparently released in this particular fission process. When 2.018 amu are added for these two neutrons, the products of the reaction have a total mass of 235.918 amu. The mass of U^{235} from the semiempirical mass formula is 235.124 amu, so that the mass of the compound nucleus $[U^{236}]$ is close to 235.124 + 1.009 = 236.133 amu. The mass excess that is converted to energy is equal to 236.133 amu - 235.918amu = 0.215 amu. Since one amu is equivalent to 931 Mev, the energy liberated in the process is $931 \times 0.215 = 198$ Mev. Although there are at least 30 different ways in which the nucleus can divide, the mass excess is approximately the same for all of these processes, and 200 Mev is a good value for the average amount of energy released per fission, as calculated in this way.

The predicted value of about 200 Mev can be compared with experimental values. The total amount of energy released per fission is the sum of the kinetic energy of the fission fragments, the kinetic energy of the emitted neutrons, the kinetic energy of the prompt γ -rays, and the

NUCLEAR FISSION

TABLE 19-6

The Energy Release in the Fission of U^{235} by Thermal Neutrons

Kinetic energy of the fission fragments	167 Mev
Kinetic energy of fission neutrons	5
Prompt γ-rays	7
β-decay energy	5
γ -decay energy	5
Neutrino energy	11
Total fission energy	200 Mev

total energy of the decay processes in the fission decay chains. It was shown in Section 19-4 that the average value of the total kinetic energy of the fission fragments from the thermal fission of U²³⁵ is 167 Mev; the uncertainty in this number is about 5 Mey. The average value of the kinetic energy carried off by the neutrons is equal to the product of the average number of neutrons emitted per fission and the average kinetic energy of the neutrons; for the thermal fission of U^{235} , this product is 2.5×2.0 Mev = 5 Mev. The kinetic energy of the prompt γ -rays is in the neighborhood of 5 to 8 Mev.^(46,47) Finally, the average energy⁽⁴⁸⁾ of all radiations (β -rays, γ -rays, and neutrinos) of the fission products is 21 ± 3 MeV; about half of this energy escapes in the form of neutrinos, and the other half is divided, approximately equally, between the β -rays and the γ -rays. These results are summarized in Table 19-6: the total fission energy determined in this way is 200 Mev, with an uncertainty between 5 and 10 Mev, in good agreement with the calculated values.

The energy release in fission has also been measured calorimetrically,⁽⁴⁹⁾ in a way which did not include the energy carried off by γ -rays, neutrons, and neutrinos. The result obtained was 177 \pm 5 Mev; if 26 Mev are added (cf. Table 19–6) for the energies missed, the total is 203 \pm 8 Mev, in good agreement with the previous results. Another calorimetric measurement yielded a value of the energy released in the fission of U²³⁵ except for the energy carried off by neutrinos;⁽⁵⁰⁾ the result was 190 \pm 5 Mev, which also agrees very well with the results of Table 19–6 and with the calculated values.

Results obtained with other fissile materials, such as Pu^{239} and U^{233} , are similar to those for U^{235} .

19-8 The theory of the fission process. It is not surprising, in view of the remarkable properties of the fission process, that a great deal of

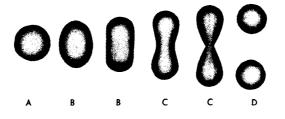


FIG. 19-8. Possible steps in the process of nuclear fission according to the liquid-drop model.

effort has been put into theoretical studies of the process. The first thorough treatment was that of Bohr and Wheeler⁽⁵¹⁾ who accounted for many of the properties of fission on the basis of the liquid-drop model of the nucleus. This model was used in the last section in calculating the energy release in fission from the atomic masses of the fissile and product nuclei as given by the semiempirical mass formula; that formula was derived by considering that a charged nucleus is analogous to a liquid drop (Section 17-6). Bohr and Wheeler showed that the liquid-drop model could be used to describe the process of fission in some detail, and made successful predictions about the existence of spontaneous fission, and about the ability of various heavy nuclei to undergo fission with slow or fast neutrons.

In the spherical, liquid-drop nucleus, the shape of the drop depends on a balance involving the surface tension forces and the Coulomb repulsive forces. If energy is added to the drop, as in the form of the excitation energy resulting from the capture of a slow neutron, oscillations are set up within the drop; these tend to distort the spherical shape so that the drop may become ellipsoidal in shape. The surface tension forces tend to make the drop return to its original shape, while the excitation energy tends to distort the shape still further. If the excitation energy is sufficiently large, the drop may attain the shape of a dumbbell. The Coulomb repulsive forces may then push the two "bells" apart until the dumbbell splits into two similar drops, each of which then becomes spherical in shape. The sequence of steps leading to fission is shown in Fig. 19-8. If the excitation energy is not large enough, the ellipsoid may return to the spherical shape, with the excitation energy being liberated in the form of γ -rays, and the process is one of radiative capture rather than fission.

The potential energy of the drop in the different stages of Fig. 19–8 can be calculated under the Bohr-Wheeler theory, as a function of the degree of deformation of the drop^(51,52) The form of the results is shown in Fig. 19–9, where the potential energy E is plotted against a parameter r which is a measure of the degree of deformation. At r = 0, which repre-

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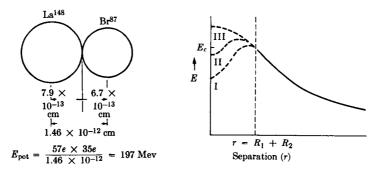


FIG. 19-9. Potential energy of two fission fragments as a function of distance between centers and the configuration at contact, where the potential energy is 197 Mev.

sents the initial spherical nuclear drop, there is available an amount of energy E_0 , given by

$$E_0 = c^2[M(A, Z) - M(A_1, Z_1) - M(A_2, Z_2)], \qquad (19-6)$$

where (A, Z) represents the nucleus that may undergo fission, and the subscripts 1 and 2 stand for the possible final products. The value of E_0 corresponds to the ground state of the compound nucleus formed when the target nucleus captures a neutron, but does not include the excitation energy resulting from the neutron capture. It was shown in the last section that the value of E_0 for U²³⁶ is about 200 Mev.

The meaning of the graph can now be seen more easily if the process which would be the reverse of fission is considered. When the drop has split into two fragments, r is the distance between their centers; if R_1 and R_2 are the radii of the two product drops, $r = R_1 + R_2$ is the value of the deformation parameter for which the two drops just touch. For values of r smaller than $R_1 + R_2$, r represents the degree of deviation of the original drop from its spherical shape. For values of r greater than $R_1 + R_2$, the energy is just the electrostatic energy resulting from the mutual repulsion of the two positively charged nuclear fragments. The value of E in this region is $(Z_1Z_2e^2/r)$, and E increases as the distance between the two fragments is decreased; the energy at $r = \infty$ is taken equal to zero. When the two fragments just touch, at $r = R_1 + R_2$, the Coulomb energy is denoted by E_c and is

$$E_{c} = \frac{Z_{1}Z_{2}e^{2}}{R_{1} + R_{2}}.$$
 (19-7)

When r is smaller than $R_1 + R_2$, the energy depends not only on the electrostatic forces, but also on the surface tension forces; the calculation

of E is then complicated, and three different curves (I, II, and III) are shown for the possible variation of E in this region. The shapes of these curves and the values of E_0 are related to the mass of the nucleus, that is, to the value of A in Eq. (19-6). Stable nuclei with values of A somewhat greater than 100 are of type I, with E_0 about 50 Mev smaller than E_c . Nuclei like those of uranium, plutonium, or thorium are of type II, for which $E_c - E_0$ is about 6 Mev. For still heavier nuclei, E_0 may be greater than E_c , as in case III. Nuclei of type III should undergo fission spontaneously and would not be expected to last long.

On a classical basis, nuclei of type II should be stable with respect to fission, but according to quantum mechanics, there is a certain probability that they will undergo spontaneous fission. The fragments may "leak through" the barrier represented by E_c in the same way that α -particles do in α -decay. In U²³⁸, about 25 spontaneous fissions occur per gram per hour, and the half-life for this process is about 10¹⁷ years.

The activation energy $E_c - E_0$ needed to induce fission in nuclei of type II has been calculated from the Bohr-Wheeler theory, and can be compared with the excitation energy resulting from the capture of a neutron, or other particle, or a γ -ray. The excitation energy of a compound nucleus can be computed with the aid of the semiempirical binding energy (or mass) formula, or from the masses of the nuclei involved if they are known. When the compound nucleus is formed by the capture of a slow neutron, the excitation energy differs by a negligible amount from the binding energy of the last neutron. In the case of [U²³⁶], the binding energy is calculated from the known masses as follows:

Mass of U^{235} ,		235.11704			
Mass of neutron,		1.00898			
		236.12602			
Mass of U^{236}		236,11912			
	$\Delta M =$	0.00690	amu =	= 6.4	Mev

The values of the excitation energy calculated in this way for a number of heavy nuclei are listed in Table 19–7 and compared with the corresponding values of the activation energy obtained from the Bohr-Wheeler theory. In U^{233} , U^{235} , and Pu^{239} the excitation energy is considerably greater than the activation energy, and these nuclei would be expected to undergo fission with thermal neutrons; they do, and the thermal fission cross sections of U^{235} and Pu^{239} are large, 577 b and 742 b, respectively. In U^{238} and Th^{232} , the excitation energy is smaller than the activation energy and these nuclei do not undergo fission with thermal neutrons, as predicted. To induce fission, the neutrons must have considerable kinetic

19-8]

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TABLE 19-7

Target	Compound	Excitation	Activation
nucleus	nucleus	energy, Mev	energy, Mev
U ²³³	[U ²³⁴]	6.6	4.6
U ²³⁵	[U ²³⁶]	6.4	5.3
U ²³⁸	[U ²³⁹]	4.9	5.5
Th ²³²	[Th ²³³]	5.1	6.5
Pa ²³¹	[Pa ²³²]	5.4	5.0
Np ²³⁷	[Np ²³⁸]	5.0	4.2
Pu ²³⁹	[Pu ²⁴⁰]	6.4	4.0

FISSIONABILITY OF HEAVY NUCLEI WITH THERMAL NEUTRONS

energy, and fission should be a threshold reaction in these nuclei, as it is. In Pa^{231} and Np^{237} , the excitation energy is greater than the activation energy, and these nuclides should undergo fission with thermal neutrons. Early experiments indicated that they do not; recent measurements show that there is some thermal fission, but the cross sections are very small (about 10^{-2} b) compared with those of U^{235} and Pu^{239} . The fact that the calculated values of the excitation energy are greater than those of the activation energy for Pa^{231} and Np^{237} does not agree with the very low probabilities of fission by thermal neutrons.

The values of the excitation energy listed in Table 19-7 indicate that the binding energy of the neutron added to a nucleus with an odd number of neutrons $(U^{233}, U^{235}, Pu^{239})$ is greater than that for a nucleus with an even number of neutrons (Th²³², U²³⁸, Pa²³¹, Np²³⁷). It is found generally that the excitation energy is greater for target nuclei with odd numbers of neutrons than for nuclei with even numbers of neutrons, and this effect is related to the presence of the "odd-even" term in the binding energy Fission with thermal neutrons occurs much more often in formula. nuclei with an odd number of neutrons than in nuclei with an even number of neutrons, as is shown by the values of the fission cross section listed in Tables 19-1 and 19-2. Nearly all of the target nuclei with large values of the cross section for fission by thermal neutrons have odd numbers of neutrons, while nearly all of the nuclei with very small cross sections have even numbers of neutrons. There is, therefore, some correlation between the values of the excitation energy and the occurrence of thermal fission, in qualitative agreement with the predictions of the Bohr-Wheeler theory. There are some obvious exceptions; 96 Cm²⁴⁰ has a very large cross section $(\sim 20.000 \text{ b})$, and ${}_{92}U^{230}$ and ${}_{92}U^{232}$ have appreciable cross sections (25 b and 80 b, respectively); each of these nuclei has an even number of neutrons. The nuclides Pa²³¹ and Np²³⁷ have low excitation energies, corresponding to their even numbers of neutrons, but their activation energies are still lower, so that they should undergo fission with slow neutrons in spite of their odd neutron numbers. The theory based on the liquiddrop model gives incorrect values of the activation energy in these cases.

The above discussion indicates that the liquid-drop model of the nucleus has been applied with some success to the problem of nuclear fission. The theory gives a qualitative, and sometimes quantitative, picture of the process and has made possible the description of some of the properties of fission, but it also has some serious defects in addition to those already mentioned. According to the theory, the most probable mode of division of the liquid-drop nucleus is into two equal fragments, so that it fails to account for the observed asymmetry of the process; it also yields values for photofission thresholds and for spontaneous fission rates which are wrong quantitatively, and whose variations from nuclide to nuclide do not agree with experiment.

Attempts have been made to modify the liquid-drop theory of fission in order to overcome these difficulties. The application of the shell model has been suggested,⁽⁵³⁾ and has had some success in accounting for the asymmetry.⁽⁵⁴⁾ Another promising approach seems to be that of Hill and Wheeler,⁽⁵⁵⁾ which is based on the new collective model of the nucleus, and the reader is referred to their article for a complete and detailed discussion of the mechanism of fission. Many additional papers on the theory of fission can be found in the general references listed below.

Chapter 4

Components of a nuclear reactor

There are several components common to most types of reactor:

Fuel

<u>Uranium</u> is the basic fuel. Usually pellets of uranium oxide (UO_2) are arranged in tubes to form fuel rods. The rods are arranged into fuel assemblies in the reactor core.* In a 1000 MWe class PWR there might be 51,000 fuel rods with over 18 million pellets.

* In a new reactor with new fuel a neutron source is needed to get the reaction going. Usually this is beryllium mixed with polonium, radium or other alpha-emitter. Alpha particles from the decay cause a release of neutrons from the beryllium as it turns to carbon-12. Restarting a reactor with some used fuel may not require this, as there may be enough neutrons to achieve criticality when control rods are removed.

Moderator

Material in the core which slows down the neutrons released from fission so that they cause more fission. It is usually water, but may be heavy water or graphite.

Control rods or blades

These are made with neutron-absorbing material such as cadmium, hafnium or boron, and are inserted or withdrawn from the core to control the rate of reaction, or to halt it.* In some PWR reactors, special control rods are used to enable the core to sustain a low level of power efficiently. (Secondary control systems involve other neutron absorbers, usually boron in the coolant – its concentration can be adjusted over time as the fuel burns up.) PWR control rods are inserted from the top, BWR cruciform blades from the bottom of the core.

* In fission, most of the neutrons are released promptly, but some are delayed. These are crucial in enabling a chain reacting system (or reactor) to be controllable and to be able to be held precisely critical.

Coolant

A fluid circulating through the core so as to transfer the heat from it. In light water reactors the water moderator functions also as primary coolant. Except in BWRs, there is secondary coolant circuit where the water becomes steam. (See also later

section on primary coolant characteristics.) A PWR has two to four primary coolant loops with pumps, driven either by steam or electricity – China's Hualong One design has three, each driven by a 6.6 MW electric motor, with each pump set weighing 110 tonnes.

Pressure vessel or pressure tubes

Usually a robust steel vessel containing the reactor core and moderator/coolant, but it may be a series of tubes holding the fuel and conveying the coolant through the surrounding moderator.

Steam generator

Part of the cooling system of pressurised water reactors (PWR & PHWR) where the high-pressure primary coolant bringing heat from the reactor is used to make steam for the turbine, in a secondary circuit. Essentially a heat exchanger like a motor car radiator.* Reactors have up to six 'loops', each with a steam generator. Since 1980 over 110 PWR reactors have had their steam generators replaced after 20-30 years service, over half of these in the USA.

* These are large heat exchangers for transferring heat from one fluid to another – here from highpressure primary circuit in PWR to secondary circuit where water turns to steam. Each structure weighs up to 800 tonnes and contains from 300 to 16,000 tubes about 2 cm diameter for the primary coolant, which is radioactive due to nitrogen-16 (N-16, formed by neutron bombardment of oxygen, with half-life of 7 seconds). The secondary water must flow through the support structures for the tubes. The whole thing needs to be designed so that the tubes don't vibrate and fret, operated so that deposits do not build up to impede the flow, and maintained chemically to avoid corrosion. Tubes which fail and leak are plugged, and surplus capacity is designed to allow for this. Leaks can be detected by monitoring N-16 levels in the steam as it leaves the steam generator.

Containment

The structure around the reactor and associated steam generators which is designed to protect it from outside intrusion and to protect those outside from the effects of radiation in case of any serious malfunction inside. It is typically a metre-thick concrete and steel structure.

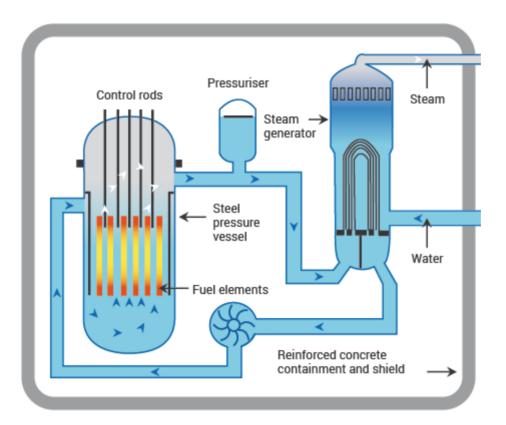
Newer Russian and some other reactors install core melt localisation devices or 'core catchers' under the pressure vessel to catch any melted core material in the event of a major accident.

There are several different types of reactor as indicated in the following table.

types of nuclear reactor

Pressurised water reactor (PWR)

This is the most common type, with about 300 operable reactors for power generation and several hundred more employed for naval propulsion. The design of PWRs originated as a <u>submarine power plant</u>. PWRs use ordinary water as both coolant and moderator. The design is distinguished by having a primary cooling circuit which flows through the core of the reactor under very high pressure, and a secondary circuit in which steam is generated to drive the turbine. In Russia these are known as VVER types – water-moderated and -cooled.



A PWR has fuel assemblies of 200-300 rods each, arranged vertically in the core, and a large reactor would have about 150-250 fuel assemblies with 80-100 tonnes of uranium.

Water in the reactor core reaches about 325°C, hence it must be kept under about 150 times atmospheric pressure to prevent it boiling. Pressure is maintained by steam in a pressuriser (see diagram). In the primary cooling circuit the water is also the moderator, and if any of it turned to steam the fission reaction would slow down. This negative feedback effect is one of the safety features of the type. The secondary shutdown system involves adding boron to the primary circuit.

The secondary circuit is under less pressure and the water here boils in the heat exchangers which are thus steam generators. The steam drives the turbine to produce electricity, and is then condensed and returned to the heat exchangers in contact with the primary circuit.

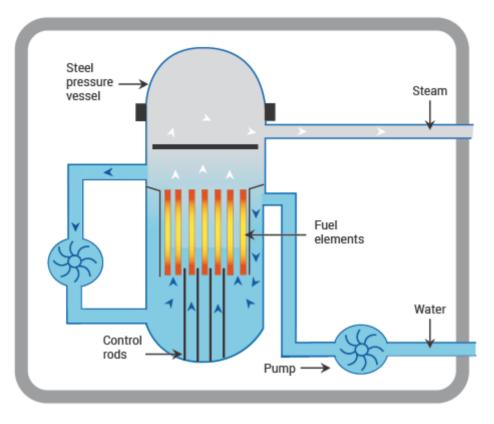
Boiling water reactor (BWR)

This type of reactor has many similarities to the PWR, except that there is only a single circuit in which the water is at lower pressure (about 75 times atmospheric pressure) so that it boils in the core at about 285°C. The reactor is designed to operate with 12-15% of the water in the top part of the core as steam, and hence with less moderating effect and thus efficiency there. BWR units can operate in load-following mode more readily than PWRs.

The steam passes through drier plates (steam separators) above the core and then directly to the turbines, which are thus part of the reactor circuit. Since the water around the core of a reactor is always contaminated with traces of radionuclides, it means that the turbine must be shielded and radiological protection provided during maintenance. The cost of this tends to balance the savings due to the simpler design. Most of the radioactivity in the water is very short-lived*, so the turbine hall can be entered soon after the reactor is shut down.

* mostly N-16, with a 7 second half-life

A BWR fuel assembly comprises 90-100 fuel rods, and there are up to 750 assemblies in a reactor core, holding up to 140 tonnes of uranium. The secondary control system involves restricting water flow through the core so that more steam in the top part reduces moderation.



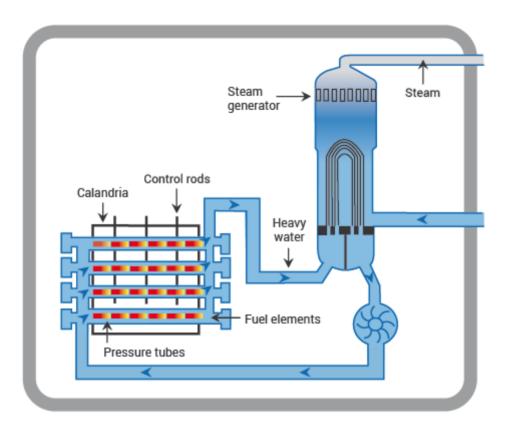
Pressurised heavy water reactor (PHWR)

The PHWR reactor has been developed since the 1950s in Canada as the CANDU, and from 1980s also in India. PHWRs generally use natural uranium (0.7% U-235) oxide as fuel, hence needs a more efficient moderator, in this case heavy water (D₂O).** The PHWR produces more energy per kilogram of mined uranium than other designs, but also produces a much larger amount of used fuel per unit output.

** with the CANDU system, the moderator is enriched (*i.e.* water) rather than the fuel – a cost tradeoff.

The moderator is in a large tank called a calandria, penetrated by several hundred horizontal pressure tubes which form channels for the fuel, cooled by a flow of heavy water under high pressure (about 100 times atmospheric pressure) in the primary cooling circuit, typically reaching 290°C. As in the PWR, the primary coolant generates steam in a secondary circuit to drive the turbines. The pressure tube design means that the reactor can be refuelled progressively without shutting down, by isolating individual pressure tubes from the cooling circuit. It is also less

costly to build than designs with a large pressure vessel, but the tubes have not proved as durable.



A CANDU fuel assembly consists of a bundle of 37 half metre long fuel rods (ceramic fuel pellets in zircaloy tubes) plus a support structure, with 12 bundles lying end to end in a fuel channel. Control rods penetrate the calandria vertically, and a secondary shutdown system involves adding gadolinium to the moderator. The heavy water moderator circulating through the body of the calandria vessel also yields some heat (though this circuit is not shown on the diagram above).

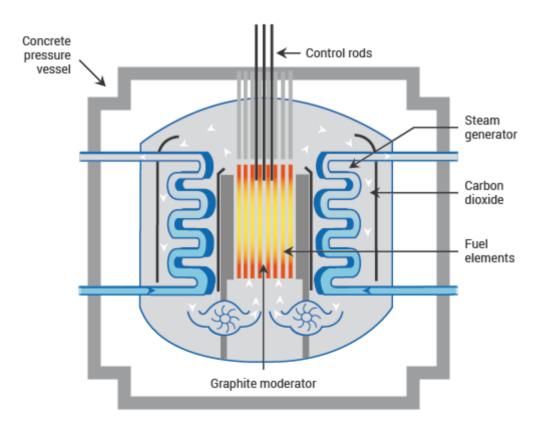
Newer PHWR designs such as the Advanced Candu Reactor (ACR) have light water cooling and slightly-enriched fuel.

CANDU reactors can accept a variety of fuels. They may be run on recycled uranium from reprocessing LWR used fuel, or a blend of this and depleted uranium

left over from enrichment plants. About 4000 MWe of PWR might then fuel 1000 MWe of CANDU capacity, with addition of depleted uranium. Thorium may also be used in fuel.

Advanced gas-cooled reactor (AGR)

These are the second generation of British gas-cooled reactors, using graphite moderator and carbon dioxide as primary coolant. The fuel is uranium oxide pellets, enriched to 2.5 - 3.5%, in stainless steel tubes. The carbon dioxide circulates through the core, reaching 650°C and then past steam generator tubes outside it, but still inside the concrete and steel pressure vessel (hence 'integral' design). Control rods penetrate the moderator and a secondary shutdown system involves injecting nitrogen to the coolant. The high temperature gives it a high thermal efficiency – about 41%. Refuelling can be on-load.



The AGR was developed from the Magnox reactor. Magnox reactors were also graphite moderated and CO_2 cooled, used natural uranium fuel in metal form, and water as secondary coolant. The UK's last Magnox reactor closed at the end of 2015.

Light water graphite-moderated reactor (LWGR)

The main LWGR design is the RBMK, a Soviet design, developed from plutonium production reactors. It employs long (7 metre) vertical pressure tubes running through graphite moderator, and is cooled by water, which is allowed to boil in the core at 290°C and at about 6.9 MPa, much as in a BWR. Fuel is low-enriched uranium oxide made up into fuel assemblies 3.5 metres long. With moderation largely due to the fixed graphite, excess boiling simply reduces the cooling and neutron absorbtion without inhibiting the fission reaction, and a positive feedback problem can arise, which is why they have never been built outside the Soviet Union. See appendix on <u>RBMK Reactors</u> for further information.

References

1 - <u>Nuclear physics by Irving Kaplan (2nd edition), 1962</u>.

2 - <u>Atomic & nuclear physics An</u> introduction 3rd edition by T.A.Littlfield & <u>N.Thorly , 1979</u>.

 $3 - \underline{\text{The physics of radiation therapy by Khan}}{\text{FM. 4}^{\text{th}} \text{ edition , 2009}}$.