S **78.** *The Reactions of High-energy Particles with Nuclei.*

By Geoffrey Wilkinson.

The use of the new 184-inch cyclotron, which affords deuterons and α-particles of much higher energy than hitherto available, has given a far more complex mixture of isotopes than the smaller cyclotrons. This has necessitated a reconsideration of the mechanisms involved in atomic fission.

The 184-inch cyclotron at the Radiation Laboratory, University of California, Berkeley, has, since early 1947, been producing deuterons of energy about 190 Mev. and α-particles of 380 Mev., and has been recently modified to produce also 350-Mev. protons. The reaction of such high-energy particles with matter produces a vast array of radioactive isotopes of elements, often far removed from the target nucleus, and it has been with these isotopes, and the mechanism of their production, that the chemistry section under Prof. G. T. Seaborg has been concerned. Since particles of energies only about one-tenth of those produced by this cyclotron were previously available, the complex reactions produced by it have had to depend on additional work at low energies using the 60-inch cyclotron for detailed interpretation. The research of Prof. Seaborg's group can be separated into three main categories.

(*a*) *The production and characterisation of new radioactive isotopes.* The high-energy particles provide an easy method of producing radioactive isotopes far on the neutron-deficient side of stability by reactions such as *d*, 6*n*; *d*, 2*p*; *d*, 5*n*, etc. Particles of energies below the available maximum can be obtained from the 184-inch cyclotron by adjusting the radius at which the probe target is set in the cyclotron. The mass allocation of new isotopes is therefore possible by excitation studies over a wide range of bombarding energies using both the 60-inch and the 184-inch cyclotron. New radioactive isotopes of elements between praseodymium and mercury have been studied by Wilkinson and Hicks (cf. preceding paper), whilst neutron-deficient radioactive isotopes of antimony, tellurium, rhodium, palladium, and cerium have been studied by Perlman, Lindner, and Chubbock (*Physical Rev.*, 1948, **73**, 1124, 1202; **74**, 982) using the 184-inch cyclotron.

In the region of the heavy elements, bombardment of uranium and thorium with high-energy particles has led to the recognition of many new α-particle-emitting isotopes, and six new "collateral" α-particle decay series (Ghiorso, Meincke, and Seaborg, *ibid.*, **74**, p. 695; Meincke, Ghiorso, and Seaborg, *ibid.*, 1949, **75**, 314) associated with the parent isotopes.

(*b*) *The study of spallation reactions of high-energy particles.* The bombardment of targets of various elements led to the observation of nuclear reactions where the product nuclei may be 20 mass units or more lighter than the target nucleus. These reactions, involving the emission of many light particles, have been quantitatively investigated by use of copper, arsenic, and uranium targets; preliminary results of some of the work have been published (Hopkins and Cunningham, *ibid.*, 1948, **73**, 1406). The term "spallation reaction" from the verb *to spall*, splinter, or chip, has been adopted for reactions of high-energy particles where multiple emission of neutrons, protons, or light nuclei occurs.

(*c*) *The study of the fission of heavy elements and the mechanisms involved at high bombarding energies.* Nuclear fission has been observed with tantalum, platinum, thallium, lead, and bismuth (Perlman, Goeckmann, Templeton, and Howland, *ibid.*, 1947, **72**, 352), whilst more detailed studies, from the mechanism standpoint, have been made with bismuth (Goeckmann and Perlman, *ibid.*, 1948, **73**, 1127) and uranium (O'Connor and Seaborg, *ibid.*, 1948, **74**, 1189).

Further, an observation, that with 380-Mev. α-particle bombardment of lead, astatine isotopes are formed in yields greater than those to be expected from reactions with impurities has been made by Templeton and Perlman. Such reactions are attributed either to secondary reactions of lithium nuclei formed by the high-energy particles, or to reactions involving emission of mesons of the type (α-uxn).

It is the characteristic spallation reactions of high-energy particles, and the correlation of experimental results with theories of high-energy-particle reactions, that will be further discussed here. The theory of nuclear reactions of high-energy protons and neutrons, which is quite different from the well-known Bohr–Wheeler theory for low-energy nuclear reactions, has been discussed by Serber (*ibid.*, 1947, **72**, 1114). In the Bohr–Wheeler picture, the projectile forms with the target a "compound nucleus" which then proceeds to dissipate the excitation energy by emission of neutrons or protons, a process more probable than emission of γ-radiation since it can occur much more quickly; an analogy has been made to the formation of, and evaporation from, a liquid drop. The break up or dissipation of excess energy from the compound nucleus is independent of the method of formation, and for a particular product the yields are highest when the excitation energy is most appropriate for the evaporation of the requisite number of neutrons or changed particles. The yield decreases rapidly with increasing energy since the probability of evaporation of further particles increases. In α-particle bombardments, for example, the yields of αn, $\alpha 2n$, $\alpha 3n$, etc., reactions rise from a threshold to a maximum, then fall off rapidly, in succession, first αn, then $\alpha 2n$, etc. On the Bohr–Wheeler theory, therefore, it might be expected for high-energy-particle reactions that no products close to the target nucleus would be produced—since the binding energy of a nucleon is roughly 8 Mev. for medium heavy elements, bombardment with 100-Mev. protons might be expected to give products about 10—12 mass units lighter than the target. That a new approach to the theory of high-energy reactions is required is immediately apparent from the fact that the highest yields of product nuclei are found within a few mass units of the target—hence, some mechanism must be found for obtaining quite small excitation energies with bombarding particles such as 180-Mev. deuterons.

According to Serber's theory, the picture is quite different from the Bohr–Wheeler picture. Considering bombardment by a single neutron or proton of high energy, the situation can be summarised as follows.

(i) The collision time between the incident particle and the target is short compared to the time between the collisions of the nucleons within the nucleus. Thus the first step in the reaction can be regarded in terms of collisions between the incident particle and an individual nucleon. The situation is not exactly that of collisions between free nucleons because of the binding of the struck nucleon within the target nucleus. From this consideration, two facts arise : (*a*) As the energy of the incident particle increases, the scattering cross-section decreases inversely. Thus the mean free path of the projectile crossing nuclear matter increases with increasing energy, *i.e.*, the nucleus begins to be transparent to the incident particle. (*b*) The incident particle loses only a small fraction of its energy to the struck one.

The mean free path for a 100-Mev. projectile is estimated to be about 4×10^{-13} cm., and the kinetic energy transferred to a struck particle about 25 Mev. It is obvious that since the mean free path of the projectile in a nucleus approximates to the nuclear radius, the effects of such a high-energy particle cannot be described in terms of the formation of a compound nucleus as is formed with particles of low energy.

(ii) The effect of a high-energy particle depends on the trajectory. If it passes near the edge of a nucleus, the particle can emerge having made only a single collision and having lost only about 25 Mev. of its energy to the target. The projectile may, of course, have changed from a proton to a neutron or *vice versa* in the process. If the projectile hits the centre—a head-on collision—multiple collisions can occur within the nucleus, which can then receive up to the full energy of the projectile in excitation energy.

(iii) Considering, now, the struck particle, this has a much lower energy than the projectile—hence a much shorter mean free path in nuclear matter. The struck particle can hence take two courses : (*a*) It escapes from the nucleus without further collisions *only* if it is near the edge and if it is heading outwards; the struck particle then emerges with 15—20 Mev. energy. (*b*) It collides with particles in the nucleus, distributing its energy among them, and subsequent events are then similar to the compound nucleus of the Bohr–Wheeler theory—the nuclear excitation is dissipated by successive boiling off of particles, each with a few Mev. energy.

(iv) The effects of a deuteron or α-particle bombardment are in effect, those of simultaneous bombardment by individual nucleons comprising the deuteron or α-particle, since the binding energy of nucleons within them affects mainly the spatial correlation between them.

(v) With deuterons a special case arises, which was first recognised from the production of a narrow band of high-energy neutrons in deuteron bombardment. This was explained (Serber, *ibid.*, 1947, **72**, 1009) by the proton of the deuteron striking the edge of the target nucleus and being stripped off, while the neutron proceeds on its way. An equal number of high-energy protons must be, and indeed are, similarly produced by stopping of the neutron. Absorption of the entire deuteron by a nucleus is of course possible with a much lower probability, when the full energy is available for nuclear excitation.

The theory thus gives an explanation for the wide distribution of products from high-energy bombardments resulting from the wide distribution of excitation energies of the struck nucleus. It also explains the loss of a small number of particles as well as the loss of many. Because of the several methods of excitation and the increasing transparency of the nucleus at higher energies, the excitation or yield function for a given product is different from that in the Bohr–Wheeler case. Since the probability of leaving a given excitation energy is determined only by the mean free path of the projectile, which varies slowly with the energy of the incident particle, the excitation curve varies only slowly at high energies.

The quantitative measurement of the products from reactions of high-energy particles is a problem similar to that encountered in the study of fission products, and requires chemical separations of very many elements from each other, in addition to accurate measurements of radioactivity. Product isotopes have been found extending over an atomic number range of 15 or so, and mass number range of 20—30. The difficulty of radioactive measurement is, moreover, increased by the formation of many neutron-deficient isotopes where the disintegration schemes and counting efficiencies are often obscure. With 400-Mev. α-particles on copper, for example, radioactive isotopes down to the 37-minute ^{38}Cl are formed. Since the precise manner of formation of a particular product is difficult to determine, the convention of writing reactions as ^{65}Cu (α, 6*z*, 18*a*)^{51}Mn has been adopted, where *z* and *a* are the loss in charge and in mass units respectively. The distribution of reaction products of high-energy deuterons with targets so far studied can be explained by the processes outlined by Serber's theory; *i.e.*, (*a*) in elastic collisions exciting the target nucleus to the extent of about 25 Mev., (ii) complete absorption of "stripped" protons or neutrons exciting the nucleus by about 100 Mev., (iii) formation of a compound nucleus by a complete deuteron with about 190-Mev. excitation energy, which can then boil off large numbers of light particles. The bombardment of copper with 190-Mev. deuterons is an example. The maximum yields of products are at masses 60 and 62, about three nucleons less than the target ^{63}Cu and ^{65}Cu. The radioactive isotopes here are thus produced by the inelastic scattering type of excitation. From 10 to 15 mass units below the target no significant decrease in yield is observed, and a possible second maximum is found at mass 51—53 representing about 12 nucleons from the target. Such isotopes are almost certainly formed by evaporation from a compound nucleus formed with about 100-Mev. excitation energy formed by absorption of a single proton or neutron. After about 15 mass units below the target, a sharp drop in the yield of activities occurs, and continues to fall until at about 30 mass units from the target, the yield is only about one-hundredth of that at 15 units below. The absorption of the complete deuteron leading to a compound nucleus with the full excitation energy, and therefore capable of evaporating large numbers of particles, is the most probable explanation of this observation. This contention is supported by the fact that bombardment of copper to 190-Mev. deuterons and 190-Mev. α-particles gives the same yield of ^{38}Cl, whereas with 380-Mev. α-particles, the yield is higher by a factor of 6.

The same general features have been observed in other cases—about 80% of reactions leading to products within a few mass units of the target, followed by comparatively high yields to about 15 mass units from the target, then a very rapid decrease in yield. The mechanisms of emission of particles are not explicitly understood. With increasing mass of the target nucleus, the neutron to proton ratio, of course, increases, and the increasing yields of neutron-deficient isotopes when the heavier elements are bombarded can be explained by primary emission of neutrons. The emission of charged particles from heavier nuclei, is, naturally, less probable than with lighter nuclei because of the charge and potential barrier. Emission of α-particles rather than neutrons plus protons is energetically more economical because of the binding energy of the α-particle; α-particle emission increases the neutron to proton ratio of the products, and may account for the increased yields of neutron-excess isotopes in the reactions with elements of lower atomic number. That α-particle emission is an important mode of energy dissipation, possibly through chains of short-lived α-particle emitters following the primary emission of neutrons from the target, is shown by the formation of ^{211}At in the bombardment of uranium with deuterons of energy as low as 50 Mev.; the formation of astatine is possible in no other way. The formation of "shielded" radioactive isotopes is a further indication of α-particle emission. It may prove possible to detect short-lived α-particle emitters of elements in the middle of the Periodic System, although competition from orbital-electron capture may be severe; an α-particle-emitting gold isotope has been recently observed by S. Thompson in the bombardment of gold with 190-Mev. deuterons, and many other such isotopes may be found.

Summarising, therefore—the main mechanisms of particle emission are (*a*) emission of neutrons with subsequent decay of the primary product nucleus by particle emission or by orbital electron capture or positron-emitter chains, (*b*) less probable, except possibly in the lighter elements, direct emission of charged particles.

The multiple emission of neutrons as a first step in the reaction of high-energy particles is also an essential feature of the fission of bismuth and other elements with high-energy particles. The results obtained by Goeckmann and Perlman (*loc. cit.*) are consistent with emission of neutrons until a nucleus is formed where the Bohr–Wheeler parameter Z^2/A is comparable to that of ^{236}U. For ^{208}Bi with deuterons such a nucleus would be ^{198}Po. Fission appears to be entirely symmetrical, and the primary fission products formed have a neutron to proton ratio which is constant, and equal to that of the fissioning nucleus, ^{198}Po. The fissioning nucleus, is, in spite of prior loss of neutrons, in an energy state far more excited than the compound nuclei formed in thermal neutron fission, *e.g.*, of ^{235}U, and hence splits exceedingly rapidly before re-arrangement to the most energetically favoured distribution of nucleons can occur; fission thus is symmetric with a constant neutron to proton ratio of the products.

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