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Artificial radioactivity produced by neutron bombardment

Nobel Lecture, December 12, 1938

Although the problem of transmuting chemical elements into each other is much older than a satisfactory definition of the very concept of chemical element, it is well known that the first and most important step towards its solution was made only nineteen years ago by the late Lord Rutherford, who started the method of the nuclear bombardments. He showed on a few examples that, when the nucleus of a light element is struck by a fast α-particle, some disintegration process of the struck nucleus occurs, as a consequence of which the α-particle remains captured inside the nucleus and a different particle, in many cases a proton, is emitted in its place. What remains at the end of the process is a nucleus different from the original one; different in general both in electric charge and in atomic weight.

The nucleus that remains as disintegration product coincides sometimes with one of the stable nuclei, known from the isotopic analysis; very often, however, this is not the case. The product nucleus is then different from all "natural" nuclei; the reason being that the product nucleus is not stable. It disintegrates further, with a mean life characteristic of the nucleus, by emission of an electric charge (positive or negative), until it finally reaches a stable form. The emission of electrons that follows with a lag in time the first practically instantaneous disintegration, is the so-called artificial radioactivity, and was discovered by Joliot and Irene Curie at the end of the year 1933.

These authors obtained the first cases of artificial radioactivity by bombarding boron, magnesium, and aluminium with α-particles from a polonium source. They produced thus three radioactive isotopes of nitrogen, silicon and phosphorus, and succeeded also in separating chemically the activity from the bulk of the unmodified atoms of the bombarded substance.

The neutron bombardment

Immediately after these discoveries, it appeared that α-particles very likely did not represent the only type of bombarding projectiles for producing

artificial radioactivity. I decided therefore to investigate from this point of view the effects of the bombardment with neutrons.

Compared with α-particles, the neutrons have the obvious drawback that the available neutron sources emit only a comparatively small number of neutrons. Indeed neutrons are emitted as products ofnuclear reactions, whose yield is only seldom larger than 10^{-4}. This drawback is, however, compensated by the fact that neutrons, having no electric charge, can reach the nuclei of all atoms, without having to overcome the potential barrier, due to the Coulomb field that surrounds the nucleus. Furthermore, since neutrons practically do not interact with electrons, their range is very long, and the probability of a nuclear collision is correspondingly larger than in the case of the α-particle or the proton bombardment. As a matter of fact, neutrons were already known to be an efficient agent for producing some nuclear disintegrations.

As source of neutrons in these researches I used a small glass bulb containing beryllium powder and radon. With amounts of radon up to 800 millicuries such a source emits about 2×10^{7} neutrons per second. This number is of course very small compared to the yield of neutrons that can be obtained from cyclotrons or from high-voltage tubes. The small dimensions, the perfect steadiness and the utmost simplicity are, however, sometimes very useful features of the radon + beryllium sources.

Nuclear reactions produced by neutrons

Since the first experiments, I could prove that the majority of the elements tested became active under the effect of the neutron bombardment. In some cases the decay of the activity with time corresponded to a single mean life; in others to the superposition of more than one exponential decay curve.

A systematic investigation of the behaviour of the elements throughout the Periodic Table was carried out by myself, with the help of several collaborators, namely Amaldi, d'Agostino, Pontecorvo, Rasetti, and Segré. In most cases we performed also a chemical analysis, in order to identify the chemical element that was the carrier of the activity. For short living substances, such an analysis must be performed very quickly, in a time of the order of one minute.

The results of this first survey of the radioactivities produced by neutrons can be summarized as follows: Out of 63 elements investigated, 37 showed

an easily detectable activity; the percentage of the activatable elements did not show any marked dependence on the atomic weight of the element. Chemical analysis and other considerations, mainly based on the distribution of the isotopes, permitted further to identify the following three types of nuclear reactions giving rise to artificial radioactivity :

$$ {}^{M}_{Z}A + {}^{1}_{0}n = {}^{M-3}_{Z-2}A + {}^{4}_{2}He \qquad (1) $$

$$ {}^{M}_{Z}A + {}^{1}_{0}n = {}^{M}_{Z-1}A + {}^{1}_{1}H \qquad (2) $$

$$ {}^{M}_{Z}A + {}^{1}_{0}n = {}^{M+1}_{Z}A \qquad (3) $$

where ${}^{M}_{Z}A$ is the symbol for an element with atomic number Z and mass number M; n is the symbol of the neutron.

The reactions of the types (1) and (2) occur chiefly among the light elements, while those of the type (3) are found very often also for heavy elements. In many cases the three processes are found at the same time in a single element. For instance, neutron bombardment of aluminium that has a single isotope ^{27}Al, gives rise to three radioactive products: ^{24}Na, with a half-period of 15 hours by process (1); ^{27}Mg, with a period of 10 minutes by process (2); and ^{28}A1 with a period of 2 to 3 minutes by process (3).

As mentioned before, the heavy elements usually react only according to process (3) and therefore, but for certain complications to be discussed later, and for the case in which the original element has more than one stable isotope, they give rise to an exponentially decaying activity. A very striking exception to this behaviour is found for the activities induced by neutrons in the naturally active elements thorium and uranium. For the investigation of these elements it is necessary to purify first the element as thoroughly as possible from the daughter substances that emit β-particles. When thus purified, both thorium and uranium emit spontaneously only α-particles, that can be immediately distinguished, by absorption, from the β-activity induced by the neutrons.

Both elements show a rather strong, induced activity when bombarded with neutrons; and in both cases the decay curve of the induced activity shows that several active bodies with different mean lives are produced. We attempted, since the spring of 1934, to isolate chemically the carriers of these activities, with the result that the carriers of some of the activities of uranium are neither isotopes of uranium itself, nor of the elements lighter than uranium down to the atomic number 86. We concluded that the carriers were one or more elements of atomic number larger than 92 ; we, in Rome,

use to call the elements 93 and 94 Ausenium and Hesperium respectively. It is known that O. Hahn and L. Meitner have investigated very carefully and extensively the decay products of irradiated uranium, and were able to trace among them elements up to the atomic number 96.*

It should be noticed here, that besides processes (1), (2), and (3) for the production of artificial radioactivity with neutrons, neutrons of sufficiently high energy can react also as follows, as was first shown by Heyn: The primary neutron does not remain bound in the nucleus, but knocks off instead, one of the nuclear neutrons out of the nucleus; the result is a new nucleus, that is isotopic with the original one and has an atomic weight less by one unit. The final result is therefore identical with the products obtained by means of the nuclear photoeffect (Bothe), or by bombardment with fast deuterons. One of the most important results of the comparison of the active products obtained by these processes, is the proof, first given by Bothe, of the existence of isomeric nuclei, analogous to the isomers UX_2 and UZ, recognized long since by O. Hahn in his researches on the uranium family. The number of well-established cases of isomerism appears to increase rather rapidly, as investigation goes on, and represents an attractive field of research.

The slow neutrons

The intensity of the activation as a function of the distance from the neutron source shows in some cases anomalies apparently dependent on the objects that surround the source. A careful investigation of these effects led to the unexpected result that surrounding both source and body to be activated with masses of paraffin, increases in some cases the intensity of activation by a very large factor (up to 100). A similar effect is produced by water, and in general by substances containing a large concentration of hydrogen. Substances not containing hydrogen show sometimes similar features, though extremely less pronounced.

The interpretation of these results was the following. The neutron and the

* The discovery by Hahn and Strassmann of barium among the disintegration products of bombarded uranium, as a consequence of a process in which uranium splits into two approximately equal parts, makes it necessary to reexamine all the problems of the transuranic elements, as many of them might be found to be products of a splitting of uranium.

proton having approximately the same mass, any elastic impact of a fast neutron against a proton initially at rest, gives rise to a distribution of the available kinetic energy between neutron and proton; it can be shown that a neutron having an initial energy of 10^6 volts, after about 20 impacts against hydrogen atoms has its energy already reduced to a value close to that corresponding to thermal agitation. It follows that, when neutrons of high energy are shot by a source inside a large mass of paraffin or water, they very rapidly lose most of their energy and are transformed into "slow neutrons". Both theory and experiment show that certain types of neutron reactions, and especially those of type (3), occur with a much larger cross-section for slow neutrons than for fast neutrons, thus accounting for the larger intensities of activation observed when irradiation is performed inside a large mass of paraffin or water.

It should be remarked furthermore that the mean free path for the elastic collisions of neutrons against hydrogen atoms in paraffin, decreases rather pronouncedly with the energy. When therefore, after three or four impacts, the energy of the neutron is already considerably reduced, its probability of diffusing outside of the paraffin, before the process of slowing down is completed, becomes very small.

To the large cross-section for the capture of slow neutrons by several atoms, there must obviously correspond a very strong absorption of these atoms for the slow neutrons. We investigated systematically such absorptions, and found that the behaviour of different elements in this respect is widely different; the cross-section for the capture of slow neutrons varies, with no apparent regularity for different elements, from about 10^{-24} cm^2 or less, to about a thousand times as much. Before discussing this point, as well as the dependence of the capture cross-section on the energy of the neutrons we shall first consider how far down the energy of the primary neutrons can be reduced by the collisions against the protons.

The thermal neutrons

If the neutrons could go on indefinitely diffusing inside the paraffin, their energy would evidently reach finally a mean value equal to that of thermal agitation. It is possible, however, that, before the neutrons have reached this lowest limit of energy, either they escape by diffusion out of the paraffin, or are captured by some nucleus. If the neutron energy reaches the thermal value,

one should expect the intensity of the activation by slow neutrons to depend upon the temperature of the paraffin.

Soon after the discovery of the slow neutrons, we attempted to find a temperature dependence of the activation, but, owing to insufficient accuracy, we did not succeed. That the activation intensities depend upon the temperature was proved some months later by Moon and Tillman in London; as they showed, there is a considerable increase in the activation of several detectors, when the paraffin, in which the neutrons are slowed down, is cooled from room temperature to liquid-air temperature. This experiment defi nitely proves that a considerable percentage of the neutrons actually reaches the energy of thermal agitation. Another consequence is that the diffusion process must go on inside the paraffin for a relatively long time.

In order to measure, directly at least, the order of magnitude of this time, an experiment was attempted by myself and my collaborators. The source of neutrons was fastened at the edge of a rotating wheel, and two identical detectors were placed on the same edge, at equal distances from the source, one in front and one behind with respect to the sense of rotation. The wheel was then spun at a very high speed inside a fissure in a large paraffin block. We found that, while, with the wheel at rest, the two detectors became equally active, when the wheel was in motion during the activation, the detector that was behind the source became considerably more active than the one in front. From a discussion of this experiment was deduced, that the neutrons remain inside the paraffin for a time of the order of 10^{-4} seconds.

Other mechanical experiments with different arrangements were performed in several laboratories. For instance Dunning, Fink, Mitchell, Pegram, and Segré: in New York, built a mechanical velocity selector, and proved by direct measurement, that a large amount of the neutrons diffusing outside of a block of paraffin, have actually a velocity corresponding to thermal agitation.

After their energy is reduced to a value corresponding to thermal agitation, the neutrons go on diffusing without further change of their average energy. The investigation of this diffusion process, by Amaldi and myself, showed that thermal neutrons in paraffin or water can diffuse for a number of paths of the order of 100 before being captured. Since, however, the mean free path of the thermal neutrons in paraffin is very short (about 0.3 cm) the total displacement of the thermal neutrons during this diffusion process is rather small (of the order of 2 or 3 cm). The diffusion ends when the thermal neutron is captured, generally by one of the protons, with production of a

deuteron. The order of magnitude for this capture probability can be calculated, in good agreement with the experimental value, on the assumption that the transition from a free-neutron state to the state in which the neutron is bound in the deuteron is due to the magnetic dipole moments of the proton and the neutron. The binding energy set free in this process, is emitted in the form of γ-rays, as first observed by Lea.

All the processes of capture of slow neutrons by any nucleus are generally accompanied by the emission of γ-rays : Immediately after the capture of the neutron, the nucleus remains in a state of high excitation and emits one or more γ-quanta, before reaching the ground state. The γ-rays emitted by this process were investigated by Rasetti and by Fleischmann.

Absorption anomalies

A theoretical discussion of the probability of capture of a neutron by a nucleus, under the assumption that the energy of the neutron is small compared with the differences between neighbouring energy levels in the nucleus, leads to the result that the cross-section for the capture process should be inversely proportional to the velocity of the neutron. While this result is in qualitative agreement with the high efficiency of the slow-neutron bombardment observed experimentally, it fails on the other hand to account for several features of the absorption process, that we are now going to discuss.

If the capture probability of a neutron were inversely proportional to its velocity, one would expect two different elements to behave in exactly the same way as absorbers of the slow neutrons, provided the thicknesses of the two absorbers were conveniently chosen, so as to have equal absorption for neutrons of a given energy. That the absorption obeys instead more complicated laws, was soon observed by Moon and Tillman and other authors who showed that the absorption by a given element appears, as a rule, to be larger when the slow neutrons are detected by means of the activity induced in the same element. That the simple law of inverse proportionality does not hold, was also proved by a direct mechanical experiment by Dunning, Pegram, Rasetti, and others in New York.

In the winter of 1935-1936 a systematic investigation of these phenomena was carried out by Amaldi and myself The result was, that each absorber of the slow neutrons has one or more characteristic absorption bands, usually for energies below 100 volts. Besides this or these absorption bands, the ab-

sorption coefficient is always large also for neutrons of thermal energy. Some elements, especially cadmium, have their characteristic absorption band overlapping with the absorption in the thermal region. This element absorbs therefore very strongly the thermal neutrons, while it is almost transparent to neutrons of higher energies. A thin cadmium sheet is therefore used for filtering the thermal neutrons out of the complex radiation that comes out of a paraffin block containing a neutron source inside.

Bohr and Breit and Wigner proposed independently to explain the above anomalies, as due to resonance with a virtual energy level of the compound nucleus (i.e. the nucleus composed of the bombarded nucleus and the neutron). Bohr went much farther in giving also a qualitative explanation of the large probability for the existence of at least one such level, within an energy interval of the order of magnitide of 100 volts corresponding to the energy band of the slow neutrons. This band corresponds, however, to an excitation energy of the compound nucleus of many million volts, representing the binding energy of the neutron. Bohr could show that, since nuclei, and especially heavy nuclei, are systems with a very large number of degrees of freedom, the spacing between neighbouring energy levels decreases very rapidly with increasing excitation energy. An evaluation of this spacing shows that whereas for low excitation energies the spacing is of the order of magnitude of 10^5 volts, for high excitation energies, of the order of ten million volts, it is reduced (for elements of mean atomic weight) to less than one volt. It is therefore a very plausible assumption that one (or more) such level lies within the slow-neutron band, thus explaining the large frequency of the cases in which absorption anomalies are observed.

Before concluding this review of the work on artificial radioactivity produced by neutrons, I feel it as a duty to thank all those who have contributed to the success of these researches. I must thank in particular all my collaborators that have already been mentioned; the Istituto di Sanità Pubblica in Rome and especially Prof. G. C. Trabacchi, for the supply of all the many radon sources that have been used; the Consiglio Nazionale delle Richerche for several grants.