

## Role of Chemistry in the Study of Atomic Transmutation

By Prof. F. A. Paneth

“THE history of Alchemy is the history of an error.” Such was, about seventy years ago, Hermann Kopp’s short summary of centuries of attempts to transmute chemical elements<sup>1</sup>. It is well known that two generations later alchemy became a reality, when it had passed from the realm of the chemists to that of the physicists, who not only possessed the right weapons for attacking the atoms but also invented methods of detecting transmutations of matter on an infinitely smaller scale than those sought by chemists. One must, however, not overlook the importance of the help which was, and is, given by chemistry in many cases where a process of transmutation has to be investigated.

A combination of physical and chemical methods, frequently manifested by the collaboration of a physicist and chemist, was a decisive factor in the early days of radioactivity. The discovery, in 1898, of the new elements polonium and radium was due to the first application, by Pierre and Marie Curie, of the methods of ‘radiochemistry’; that is, the ordinary methods of analytical chemistry were employed with invisible substances, and the effect of the attempted separations controlled by electrometric apparatus for the measurement of radioactive rays. Soon afterwards the ‘active deposits’, first considered as a sort of ‘induced activity’, were recognised as chemical substances with a definite behaviour through their response to treatment with acids, co-precipitation with various metals, etc. Further, the theoretical foundation of the whole science of radioactivity, the disintegration theory of Rutherford and Soddy, could only be laid after uranium and uranium X, thorium and the rare gas thorium emanation—to quote only two of the historically most important cases—had been chemically separated and their isolated radiations investigated.

After a few years of such radio-chemical studies, chemistry, making use only of its familiar methods, was able to confirm the fact of radioactive transmutation. A natural consequence of Rutherford’s recognition of the  $\alpha$ -rays as charged helium atoms was the expectation that any radio-element which emitted  $\alpha$ -rays would evolve in the course of time a definite amount of helium gas. When in 1903 Ramsay and Soddy<sup>2</sup> succeeded in separating, purifying and identifying spectroscopically the helium newly formed in a radium salt, this first chemical demonstration of the fact that a well-

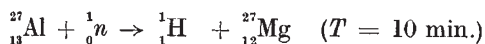
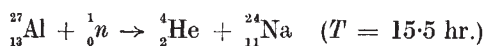
known element had obviously sprung from another well-defined chemical element appealed especially to all those not acquainted with the radio-chemical technique of identifying invisible substances; although, therefore, the result of this experiment had been predicted, its influence on the definite acceptance of the idea of elementary transformation was very marked. It was comparatively easy later to show in a similar fashion the evolution of radium emanation from a radium solution, and to measure its quantity<sup>3</sup> and photograph its spectrum<sup>4</sup>; but besides helium and radon there is only one more element which, as a product of radioactive disintegration in the laboratory, has ever been chemically identified: radium D, originating from radon, has been collected by electrolytic deposition on a platinum anode, where it displays the characteristic colour and electrochemical potential of the isotopic lead superoxide<sup>5</sup>.

Neither radio-chemical nor chemical methods played any part in the discovery and early development of the artificial transmutation of elements brought about by bombardment with  $\alpha$ -rays. The fluorescent screen, used by Rutherford and Chadwick in their pioneer work in 1919, as well as the Wilson chamber and electrometric devices which were applied later, were able to record one single atomic disruption; and so much information could be collected by the careful interpretation of the observed phenomena that in many cases it was possible to write the full equation of the transmutation, including the chemical character of the new products, without a single chemical experiment having been performed. In this way even new atomic species of known elements have been discovered, and atomic weights have been determined with an accuracy superior to all other figures previously obtained<sup>6</sup>. Nevertheless, if chemical, or at least radio-chemical, methods could have been applied, they would not only have offered a means of checking indirect conclusions but also in many cases even decided between different possibilities. Radio-chemical methods were, naturally, excluded so long as only stable atoms were found as the products of artificial disintegration; but chemical detection of, for example, hydrogen produced by the  $\alpha$ -ray bombardment of nitrogen, did not seem to be entirely out of the question, since the sensitivity of the spectroscopic identification of hydrogen in helium, according to a

statement of Collic and Ramsay<sup>7</sup>, should be just high enough. A recent investigation<sup>8</sup>, however, showed the experimental basis of this claim to be wrong.

The whole situation has now changed since the discovery of artificial radio-elements and the appearance of neutrons and of helium atoms among the products of artificial disintegration. When, two years ago, Irene Curie and F. Joliot<sup>9</sup> observed that in some cases the results of artificial transmutation were not stable but that they decayed with the emission of radiation much in the same way as the natural radio-elements, all the methods developed for the study of the latter became immediately applicable. While the instantaneous process of collision can only be investigated by the apparatus mentioned above, the subsequent radioactive disintegration opens the field for characterisation of the new products by their periods of decay, the quality of their rays, and—last but not least—their radio-chemical behaviour. Curie and Joliot<sup>10</sup> showed, for example, that a minute percentage of boron atoms bombarded by  $\alpha$ -rays are transformed into a substance which emits positrons, decays with a half-value period of 14 minutes, and shows the chemical reactions expected of nitrogen; on the other hand, the chemistry of a product generated in the same way from aluminium accorded with the supposition of its being phosphorus. Here, for the first time in processes of artificial transmutation, it was proved chemically that new elements had been formed, and from the positions of these elements, as compared with the old ones, the mechanism of the nuclear changes could be deduced, independently of the conclusions reached by physical observations.

This fruitful collaboration of the chemist with the physicist was continued on a much larger scale by Fermi and his co-workers<sup>11</sup>, who showed that bombardment by neutrons was effective in creating artificial radio-elements throughout the periodic system, up to the highest elements. The impact of a neutron on the nucleus of an atom may result in its capture, with or without simultaneous ejection of an  $\alpha$ -particle or a proton. In the case of aluminium it was found that all three reactions occur:



In order to demonstrate the isotopy of the three new radio-elements with sodium, magnesium and aluminium respectively, the same method could

be applied which, especially in the hands of Soddy and Fleck<sup>12</sup>, had proved so useful in establishing the chemical character of the shorter-lived natural radio-elements; small amounts of the supposed ordinary isotopes were added and, after thorough mixing, it was shown by analytical chemical operations that the activity of 15.5 hours half-period was inseparable from the sodium, that of 10 min. half-period was inseparable from the magnesium, and that of 2.3 min. half-period inseparable from the original element aluminium.

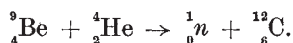
In cases like the last where isotopy with the bombarded element has to be tested, it is especially necessary first to ensure the completeness of the mixing, because it is well known that differences in the chemical or colloidal state between the radio-element and its isotope, which have been present from the beginning, may give a false impression of a chemical separability<sup>13</sup>. Now, as a consequence of the formation of the artificial radio-element by the capture of a neutron, the chemical linkage of its parent element has necessarily been broken, and the new radioactive atom may be in quite a different chemical or physical state; the fact that for this reason the new isotope can often be easily severed from the parent element, although not a 'separation of isotopes' in the technical sense, provides a valuable method for its concentration, as was first pointed out by Szilard and Chalmers<sup>14</sup>.

Other similar instances in which the study of the chemical nature of artificial disintegration products has been useful in clearing up the mechanism of the nuclear change are already much too numerous to be mentioned here. Naturally the more complicated the transformation process, so much the more welcome becomes the help of the chemist. Bombardment of thorium seems to lead to the building up of a series of subsequent radio-elements, in its general behaviour similar to, but differing from, the three known disintegration series; chemical investigation has assisted in disentangling the mixture of the new active atoms<sup>15</sup>. It has been reported<sup>16</sup>, too, that the active isotope of chlorine  ${}_{17}^{38}\text{Cl}$ , resulting from bombardment of ordinary chlorine with slow neutrons, does not immediately change into a stable substance, but that first an isotope of argon  ${}_{18}^{38}\text{A}$  is formed, which, with emission of  $\alpha$ -rays, is converted into a sulphur isotope  ${}_{16}^{34}\text{S}$ ; the inertness of the noble gas argon should make it a simple affair for the chemist to test this assumption of an active argon isotope.

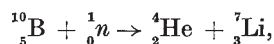
A much more difficult but very interesting task was set to chemistry after it had been found by Fermi and his collaborators that neutron bombardment of uranium produced a '13-minute-body' and a '90-minute-body', the position of which in

the system of elements could be either below or above uranium, highest of the known elements, since the mechanism of their formation remained obscure. Peculiar features in the analytical chemical behaviour made Fermi<sup>17</sup> believe that unknown elements above uranium had been formed. This was challenged by v. Grosse<sup>18</sup>, who stated that the reactions of both bodies were identical with those of element 91, protactinium; but the discoverers of protactinium, Hahn and Meitner<sup>19</sup>, by mixing the new radio-elements with the protactinium isotope uranium Z, showed them to be separable from protactinium, thus disproving Grosse's statement and supporting Fermi's claim of having discovered elements with atomic numbers higher than 92. From a study of these two bodies, which would appear to be subsequent members of a radioactive series, and perhaps themselves complex, chemistry, in its turn, will gain information about a previously inaccessible region of the periodic system.

We have mentioned above that in the development of the science of natural radioactivity, first radio-chemical, and then purely chemical, methods could be used for the identification of the products of transmutation. To-day, in the study of artificial transmutations, the second stage has been likewise reached. It is obvious that the task is here more difficult, owing to the much lower yield of newly formed matter. If we have at our disposal a quantity of 100 m.c. radon, by its complete decay  $2 \times 10^{-4}$  c.c. helium is formed; if, however, we use the same quantity of radon for the classical artificial disintegration of nitrogen, the amount of hydrogen produced is only of the order of  $10^{-9}$  c.c., as only about one in a hundred thousand  $\alpha$ -particles liberates a proton from a nitrogen nucleus; which, together with special difficulties met with in the microchemistry of hydrogen, explains the failure of the attempt mentioned above to identify by chemical means the artificially produced hydrogen. Recently, however, more effective processes of artificial transmutation have been discovered; by their impact on beryllium, one in about ten thousand  $\alpha$ -particles knocks out a neutron from a beryllium nucleus<sup>20</sup> according to the equation:



The neutrons are beyond chemical detection, nor is it possible to find such small traces of carbon in beryllium; but, after being slowed down by collision with hydrogen nuclei, the neutrons can be used up almost quantitatively by the reaction<sup>21</sup>:



thus producing an approximately equal number of helium atoms; and the microchemical detection and measurement of small quantities of helium is astonishingly safe. In an experiment, based on these considerations, the neutrons, produced by the decay of about 2,000 mc. of radon mixed with beryllium, were used for the bombardment of one litre of boron methylete; less than  $10^{-10}$  gm. of boron was disintegrated according to the above equation, and the corresponding  $10^{-7}$  c.c. of helium separated from the ester, identified spectroscopically, and measured by a special manometer<sup>22</sup>.

This fact that the artificial generation of an element has now been chemically proved by its preparation in bulk, without making use for its identification of any sort of radioactive method, such as fluorescent screen, Wilson chamber or electrometer, is a step nearer to the old alchemistic goal, although the amount of newly formed matter is still so small that only delicate microchemical methods can deal with it. As in Ramsay and Soddy's experiment, in which the production of helium by radioactive decay was shown for the first time, the qualitative result could be predicted; but in both cases the way to exact quantitative measurements has thereby been opened. It may well be that there are processes of transmutation going on with so little energy that the radio-physical and radio-chemical methods of detection cannot be applied; there is now hope that in some such cases ordinary chemistry will be able to discover the newly formed elements.

<sup>1</sup> H. Kopp, "Beiträge zur Geschichte der Chemie", I (Braunschweig, 1869), p. 17.

<sup>2</sup> W. Ramsay and F. Soddy, NATURE, 68, 246 (1903). Proc. Roy. Soc., A, 72, 204 (1903).

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<sup>4</sup> E. Rutherford and T. Royds, Phil. Mag., 16, 313 (1908).

<sup>5</sup> G. v. Hevesy and F. A. Paneth, Ber. deutsch. chem. Ges., 47, 2784 (1914).

<sup>6</sup> H. Bethe, Phys. Rev., 47, 633, 795 (1935). M. L. E. Oliphant, A. R. Kempton and Lord Rutherford, Proc. Roy. Soc., A, 150, 241 (1935).

<sup>7</sup> J. N. Collie and W. Ramsay, Proc. Roy. Soc., A, 59, 257 (1896).

<sup>8</sup> F. A. Paneth and P. L. Günther, NATURE, 131, 652 (1933); Z. phys. Chem., A, 173, 401 (1935).

<sup>9</sup> I. Curie and F. Joliot, C.R., 198, 254 (1934).

<sup>10</sup> I. Curie and F. Joliot, C.R., 198, 559 (1934).

<sup>11</sup> E. Fermi, E. Amaldi, O. D'Agostino, F. Rasetti and E. Segrè, Proc. Roy. Soc., A, 146, 483 (1934); E. Amaldi, O. D'Agostino, E. Fermi, B. Pontecorvo, F. Rasetti and E. Segrè, Proc. Roy. Soc., A, 149, 522 (1935). O. D'Agostino, Gazz. chem. ital., 64, 835 (1934).

<sup>12</sup> A. Fleck, Trans. Chem. Soc., 103, 381, 1052 (1913).

<sup>13</sup> See, for example, "Handbuch Phys.", 22 (1) (Springer, Berlin, 1933), 445.

<sup>14</sup> L. Szilard and T. A. Chalmers, NATURE, 134, 462 (1934). F. A. Paneth and J. W. J. Fay, NATURE, 135, 820 (1935).

<sup>15</sup> I. Curie, H. v. Halban and P. Preiswerk, C.R., 200, 1841 and 2079 (1935). O. Hahn and L. Meitner, Naturwiss., 23, 320 (1935).

<sup>16</sup> W. F. Libby, M. D. Peterson, W. M. Latimer, Phys. Rev., 48, 571 (1935).

<sup>17</sup> E. Fermi, NATURE, 133, 898 (1934).

<sup>18</sup> A. v. Grosse and M. S. Agruss, Phys. Rev., 46, 241 (1934); NATURE, 134, 773 (1934); J. Amer. Chem. Soc., 57, 438 (1935).

<sup>19</sup> O. Hahn and L. Meitner, Naturwiss., 23, 37, 230 (1935).

<sup>20</sup> R. Jaekel, Z. Phys., 91, 493 (1934).

<sup>21</sup> J. Chadwick and M. Goldhaber, NATURE, 135, 65 (1935); Proc. Cam. Phil. Soc., 31, 612 (1935). H. J. Taylor and M. Goldhaber, NATURE, 135, 341 (1935). E. Fermi and co-workers, Proc. Roy. Soc., A, 149, 522 (1935).

<sup>22</sup> F. A. Paneth and H. Loleit, NATURE, 136, 950 (1935).