Personal Reminiscences of a Radiochemist.

THE FARADAY LECTURE, DELIVERED BEFORE THE SOCIETY AT NOTTINGHAM ON MARCH 28TH, 1956.

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I CONSIDER it a great honour that you have invited me to give a lecture associated with the name of Michael Faraday. And when I look through the list of eminent scientists who have lectured in memory of that truly epoch-making chemist and physicist I would be ashamed to think of competing with them.

I have devoted a great many years to Science, but nearly all of them only to research on radium and other radioactive substances. What a narrow field this seems when compared with the gigantic achievements of the man whose work we are commemorating today, and very narrow too in comparison with the content of earlier Faraday lectures.

For the last ten years or more I have had no opportunity of doing any scientific research, so I have no new results of my own to submit to you. You have however very kindly allowed me to tell you something about my earlier work and it is with sincere gratitude that I avail myself of your permission to turn this Faraday Lecture into something more like a piece of personal history—a short survey of my former researches. It is also a source of peculiar gratification to me that at the close of my scientific life you have afforded me the opportunity of once more lecturing in the country in which my career as a radiochemist began, that is, in England.

It was in the autumn of 1904 that I came to London. The only asset I had was an introduction to Sir William Ramsay from Professor Zincke, the Professor in Marburg who had supervised my doctor thesis (on some branch of organic chemistry) and under whom I had subsequently worked for two years as assistant. Actually I had come to England to learn the language, because there was a prospect of my being appointed to a special post as organic chemist in the German chemical industry; and this would have entailed a certain amount of work abroad.

It was therefore pure chance—and a very lucky chance—that I had the good fortune to work under Sir William Ramsay; and it was followed by what turned out to be a whole series of lucky chances in my scientific career.

When I called on Sir William he asked me if I would like to work with radium. I told him that to date I knew nothing whatever about radium; whereupon he said that was no disadvantage, it would mean that I tackled the work without prejudice.

Sir William then handed me a bowl holding a considerable quantity of barium chloride, remarking that it contained about 10 milligrams of radium. I was to separate the radium by Madame Curie's method of fractional crystallization and produce from it a series of organic salts with the object of determining the atomic weight of radium. Owing to the high molecular weights of the salts, the 10 milligrams would possibly be sufficient for the purpose.

I started to work. I became acquainted with the transformation products of radium—its emanation and its active deposit—and checked the progress of the fractional crystallization. I obtained some strange results and very soon became aware of the appearance, side by side with the long-lived radium emanation, of a short-lived emanation which turned out to be thorium emanation. As the emanation was much more intense than that derived from thorium salts of equal weight, I was obliged to conclude that my preparations contained a new radioelement—the parent substance of this thorium emanation. I called the new element radiothorium. The explanation lay in the fact that the radium preparation which had been handed to me had been extracted from a mineral which contained both thorium and uranium. The discovery of radiothorium was therefore purely accidental.

Ramsay was interested in this result and helped me, particularly in the matter of my lack of space. I was desperately in need of some place to carry out my measurements, as Sir William Ramsay's laboratories at the Institute were very overcrowded. After a while he gave me a master-key which enabled me to enter the Chemical Institute and the neighbouring Physics Institute at night and carry out my measurements in one of its ante-rooms.

When my stay in London was drawing to a close, Sir William asked me what my future plans were. I told him that I intended to go into chemical industry. He suggested however that I should continue working on radioactivity and try for a Readership at the University of Berlin with a dissertation on this new subject. Following up that idea, Sir William wrote a letter to his friend Emil Fischer, the Director of the Berlin Chemical Institute, and Fischer replied that he was willing to take me.

This decided me to continue on the path of radium research, but I was nervous because I still knew so little about the subject. So in the summer of 1905 I wrote to Professor Ernest Rutherford in Montreal and asked him whether I might work under him for a time, that I had discovered a new radioactive element and was anxious to learn more about the subject. Rutherford sent me a very friendly answer; but he did not mention radiothorium.

When I joined him in Montreal in 1905 he questioned me about the new radio-element, but I noticed a certain reserve in his manner. After a few days he admitted to me that he had not believed in radiothorium, the discovery of which I had already briefly published. Later I heard that the eminent radiochemist B. B. Boltwood, a friend of Rutherford's, had written to him about radiothorium in a letter which contained the sentence: " The radiothorium discovered by Hahn seems to be a mixture of thorium X and stupidity." However, Boltwood was soon convinced. But the half-life value of the new substance became a subject of controversy. I had reported that the radiothorium disintegrates with a half-life period of about two years. Boltwood's thorium salts, however, showed too little activity (they did not contain the equilibrium amount of radiothorium) and this did not increase in accord with the 2 years' half-life. So my determination seemed to be wrong. When discussing the matter with him, I set up the hypothesis that it might be possible to explain our differing results by means of a longer-lived non-radiating substance between thorium and radiothorium, one which was not contained in his weak thorium salts and would be produced later.

After my return to Germany, I found that hypothetical substance; I called it mesothorium. Incidentally, the discussion which Boltwood and I had had on the subject led to a close friendship which lasted till his early death. When I wrote to him about mesothorium he congratulated me and pointed out that my hypothesis had proved correct after all.

While in Montreal, I found a new product in actinium—an intermediary substance between actinium itself and actinium X, which had been discovered under Rutherford; I called it radioactinium. It had been overlooked in Montreal and so I had my own little triumph to compensate for their disbelief in my radiothorium. In that first decade of radium research it was still very easy to discover new radio-elements.

Together with Rutherford I investigated the range of the α-rays emitted by radiothorium and its α-radiating transformation products, confirming that α-rays emitted by uniform substances have a uniform range. In the course of these researches I also found a very short-lived transformation product of the active deposit; today this is known as thorium C′.

While working with thorium preparations which led to the discovery of mesothorium in Berlin, I was supplied with very pure thorium salts. In these I found, besides the short-lived thorium emanation, very minute traces of radium emanation which increased linearly with the time. From this it was easy to deduce that the thorium contained traces of radium's parent substance, which had long been searched for, and that chemically it must be very like thorium. I started to look for this substance in the " hydroxides " group obtained in the process of extraction of radium from pitchblende. These hydroxides had to contain the thorium-like substance. But before I obtained a sample of these hydroxides from Austria, Boltwood published his discovery of ionium, radium's long-lived parent substance. This time Boltwood was ahead of me.

Boltwood too stressed the similarity between his ionium and thorium—a similarity already indicated by my observations in connection with the pure thorium salts.

About that time my attention was drawn to certain other great chemical similarities. Despite numerous attempts I was unable to separate mesothorium from radium and I very soon realized that the radiothorium in Ramsay's radium–barium salt had been generated from the mesothorium which had come down together with the radium. As mesothorium could not be separated from radium and as it did not emit any perceptible rays, I had been unable to find it earlier.

A similar lack of success having attended my efforts to separate radiothorium from thorium, it was a logical conclusion that these elements too must be chemically very much alike. But I lacked the courage to conclude that the chemical properties of, for instance, radium, mesothorium, and thorium X, or of thorium, ionium, and radiothorium were genuinely identical. Frederick Soddy, a few years later, was the first to realize that certain elements exist which have identical chemical properties and occupy the same position in the Periodic Table, but whose atomic weights differ. He called them isotopic elements. Thus, with Soddy's discovery of isotopes, all the difficulties in the way of classifying the various radio-elements vanished at one blow. Since that time we no longer speak of inseparable elements, but of isotopic atoms of one and the same element.

In the autumn of 1907 Dr. Lise Meitner, a young physicist, came from Vienna to Berlin to improve her theoretical physics under Max Planck, and also to continue the work on radioactivity that she had begun in Vienna. She came to me, and out of that visit to Berlin there grew up our collaboration and a friendship which has lasted for over thirty years.

At that time I already had quite a number of different radioactive preparations, and so Lise Meitner and I began a systematic study of the β-rays emitted by these substances. We drew up exact absorption curves, believing that uniform radio-elements would emit uniform β-rays, that is, rays of identical velocity. Working on this hypothesis we actually did find β-rays emitted by elements which till then had been considered to be non-radiating.

But when we examined the rays from very thin foils of the matter in a magnetic field—just as Rutherford had done earlier with the α-particles—it became evident that our hypothesis was wrong. We obtained line spectra, and the rays were anything but uniform. In the long run, however, work on the magnetic β-spectra led to most important results, mainly obtained by Lise Meitner and her co-workers, particularly in connection with the part played by β- and γ-rays in atomic disintegration.

To return to chemistry : I should now like to show by an example how quite unspectacular observations can often lead to interesting results.

In 1907 the Vienna Radium Institute published an article on a very slight activity observed in the active deposit of actinium, for which no explanation could be found. Could there be traces of a new radio-element ? On examination I found that the activity was caused by very small quantities of actinium X, the parent-substance of actinium emanation. But how did this definitely non-volatile alkaline-earth metal get into the active deposit ? The explanation was found by means of the following observations : Thin foils of radioactinium, the parent substance of actinium X, and equally thin foils of actinium X itself, were compared. Residual activity was found in the active deposit when the radioactinium was used, but was not found at all in the case of actinium X. It followed that the α-particles of the radioactinium were responsible for the remaining activity. The α-particles of the radioactinium cause a " recoil " of the decay product actinium X, part of which may be ejected from the preparation; in consequence, minute traces can be found outside.

A short time later Lise Meitner and I, using this new " recoil " method, had discovered some new short-lived transformation products from α-radiating precipitates. Today the recoil method is widely used in nuclear physics to separate active disintegration products. The recoil of a radioactive disintegration may lead, not only to a physical separation, but also to the breaking of chemical bonds; such a procedure is now frequently applied in the concentration of artificial radio-elements and known under the name of the Szilard–Chalmers process.

Another observation, which was at first erroneously interpreted, later led to a method used in applied radiochemistry which I called the " emanation " method.

It depends on radium and thorium emanations from precipitates which contain traces of the emanating radio-elements.

Numerous radiochemical studies were undertaken with this " emanation " method : systematic surface changes under thermal treatment; lattice transformations of crystalline substances, where the transformation is recognizable by a momentary outburst of the emanation; disintegration temperatures; and reactions in the solid state—all these can be examined by the " emanation " method. Later, Dr. Strassmann and I used this method during our work on " fission " of uranium to obtain short-lived products from xenon and krypton given off during fission by uranium preparations of very high surface area.

Once Frederick Soddy had discovered isotopes and the Rutherford–Bohr model of the atom had been set up, the system of our chemical elements became clear for all to see.

But up to the year 1917, one active element was still missing, namely the parent substance of actinium. It had to be an element with the nuclear charge 91. Indeed an isotope of this element was found by Fajans and Göhring which disintegrated in a very few minutes and which they called brevium; today it is called UX_2. But despite intensive search Fajans and Göhring did not discover the parent substance of actinium. It was found simultaneously and independently by Dr. Meitner and myself and by Soddy and Cranston. We named it protactinium. Its isolation in a pure form was first achieved by my co-worker, A. von Grosse. Later we produced several hundred milligrams of this element in the form of a double fluoride. Protactinium has a half-life of 32,000 years and can therefore be extracted by the gram from uranium minerals. The name protactinium has since been generally adopted for this long-lived form of the element 91.

In connection with element 91 I should like to give another example, this time again to show that it is important to follow up even infinitesimal inconsistencies when investigating radiochemical phenomena. Thirty years ago I noticed in the filtrate of a uranium X precipitate that its decay was not proceeding quite as one would have expected. The filtrate should have contained only the short-lived uranium X2. But in one preparation of the filtrate, I found, surprisingly, a just detectable residual activity which decreased rather too slowly. Repeating the experiment with stronger preparations, I discovered traces of the presence of an unknown radio-element with a 6·7-hour half-life; its activity was only about one-thousandth of that of uranium X and was therefore not easy to detect. It looked as if the unknown substance were a member of a hitherto undiscovered radioactive disintegration series, and I therefore tried to find the parent substance of this new body which I called uranium Z.

But these experiments led to no result and it became evident that uranium Z, like uranium X2, known much earlier, was derived from the well-known uranium X1, that is, they both had the same parent substance. In itself this was not a very spectacular result; but it happened to be the first case in which " isomerism " had been definitely proved in the course of a radioactive transformation. The next cases of nuclear isomerism were not found until twenty years later. Today they play an increasingly important part in atomic physics.

Other radiochemical investigations which we undertook at that time were of value for our later research on uranium. I refer to the detailed observations we made on the separation of minute quantities of radioactive substances with weighable amounts of precipitates. We investigated systematically the difference between the mixed-crystal type in the lattice and the adsorption of infinitesimal quantities which depended on precipitation conditions. Following experiments by Paneth and by Fajans, we formulated rules for precipitation and adsorption which clarified the behaviour of minute quantities of radioactive substances under the various conditions of precipitation. As a secondary result we found an explanation for the strange presence of helium in certain potassium salts occurring in Nature when other potassium salts from the same source contain no helium.

The discovery of protactinium, element 91, filled the last gap in the three great transformation series of the natural radio-elements.

In the early twenties, however, a new " age of nuclear research " set in. It was introduced by the first *artificial* transformation of one element into another due to the genius of Ernest Rutherford; when nitrogen nuclei were bombarded with helium nuclei, that is α-particles, the nitrogen nuclei were converted into oxygen and hydrogen. Artificial transmutation of the elements had become an accomplished fact. At first, it is true, this had only been achieved for infinitesimally minute quantities, and there was no hope that the discovery could be further exploited.

Then, in the early thirties, came the discovery of the neutron by James Chadwick, and in 1934 M. and Mme. Joliot-Curie discovered artificial radioactivity.

The discovery of the neutron meant that there was a simple explanation for isotopic phenomena. Chemical elements are composed of protons and neutrons. The number of protons determines the atomic number, that is, the chemical nature of the element. The sum of the protons and the neutrons gives the atomic weight.

In the course of atomic nuclear research it was soon realized that the uncharged neutrons are a peculiarly suitable munition for achieving atomic transformations; and it was the Italian scientist Enrico Fermi who pointed out the great importance of neutrons for nuclear transformations. He and his co-workers bombarded almost all the elements in the whole Periodic Table and produced numerous artificial radioactive isotopes. In general, the process consists in the neutron's being taken up by the nucleus; but the newly formed isotope is unstable and, emitting β-rays, is converted into the next higher element.

Fermi had carried out these experiments up to uranium and here too had found two very rapid transformations with β-ray emission. Quite logically he concluded that with uranium the process was the same as with the lower elements, and that therefore isotopes of the next higher element, that is, of element 93, had been produced from uranium, element 92.

Fermi's conclusions, however, were criticized by others. It was suggested that a 13-minute body found by Fermi might be an isotope of the next lower element 91, that is, an isotope of protactinium. As Dr. Meitner and I had found protactinium twenty years earlier and were acquainted with its chemical properties, and as moreover I had discovered uranium Z, the β-ray emitting isotope of protactinium that disintegrates in about 6 hours, we felt authorized to check Fermi's experiments. We bombarded uranium with a neutron-producing radium–beryllium preparation, added uranium Z to the solution, and with that indicator for element 91 we examined the chemical properties of Fermi's 13-minute body. The result left in us no shadow of a doubt: Fermi's discovery was certainly not element 91, so his conclusion that it was an isotope of element 93 must be correct.

It should be pointed out that at that time no other possibility was to be thought of. A large number of nuclear reactions had been demonstrated with the help of artificial radiation sources and with the neutron. The resulting products were always isotopes of the bombarded element or of its nearest neighbours in the Periodic Table. It was considered impossible that there could be any different reaction.

Together with Fritz Strassmann, Professor Meitner and I continued the experiments on the bombardment of uranium which had been begun by Fermi, because the processes turned out to be much more complicated than was at first suspected. In the course of several years we isolated a large number of artificially active atoms which developed from each other under β-radiation and were therefore taken to be transuranic elements with rising atomic numbers. We believed we had reached element 96. In so far as our experimental work was repeated in other research institutes, these results were fully confirmed.

In July 1938, Lise Meitner had to leave Germany because of Hitler's racial laws; our collaboration came to an enforced end, but our friendship endures to this day. Strassmann and I continued the experiments and, while checking some new work of Mme. Joliot-Curie and Savitch, in the autumn of 1938 we again reached some very curious results. Apart from the numerous substances which were considered to be transuranic, we found a new group of artificial atoms which, we assumed, were artificial radium isotopes. Our conclusion was inescapable insofar as the chemical properties of these substances limited the possibilities only to radium or to the barium which had been used as separator. Barium was excluded by the rules of physics as they then stood—the physicists refused even to discuss any other possibility. Therefore only radium remained.

As became clear later we were very fortunate in having only very weak preparations at our disposal. In order to measure our preparations more accurately, we had to use thinner films, particularly for a long-lived isotope with a half-life period of 300 hours with very absorbable β-rays. Therefore we had to try to separate this 300-hour substance from barium. As is well-known, this is done by fractional crystallization, a process with which we had been familiar for many years; in particular, I had used it for the concentration of the radium isotope, mesothorium, from barium. But all our experiments failed; our radium did not increase in the first crystallizations. It seemed plausible to attribute this failure to the extremely low activity of our preparations; it was possible that the very few atoms present were simply carried along, neither increasing nor decreasing.

In order to test this, we then carried out identical experiments with equally small activity, using the natural radium isotopes, mesothorium and thorium X. These substances were obtained in a state of pure radioactivity, that is, separated from the parent substances and transformation products, and subjected to the same crystallization methods as we had used with our artificial preparations. The result was the one to be expected for radium. Both mesothorium and thorium X became concentrated in the right fractions to a degree which coincided with our earlier results many years before. Here was the proof that even a very few atoms of natural radium isotopes reacted in exactly the same way as strong concentrations.

Finally we changed over to the direct " indicator experiments," first introduced into radiochemistry by Hevesy and Paneth. We mixed the pure *natural* radium isotopes with our *artificial* " radium isotopes "—previously purified from their transformation products—and proceeded with our fractional crystallization as before. *In the end products the natural radium isotopes were separated from barium, the artificial ones were not.*

Furthermore we undertook exactly analogous experiments with the direct disintegration products of our artificial radium isotopes and with the actinium isotope, mesothorium 2, obtained from mesothorium. The actinium isotopes was separable from lanthanum (which had been added as carrying agent), but the artificial transformation products were not—they were chemically identical with lanthanum. In this way we had once again ascertained that our alkaline-earth isotopes which we had till then taken to be radium isotopes were in reality artificial barium isotopes whose β-radiation produced lanthanum, but not actinium.

I need not detail the rest of these experiments. At the time of our first publication, the measurements of the indicator experiments of which I have spoken were not yet quite complete and we therefore expressed ourselves still cautiously. But by then it was already impossible that we could be mistaken.

That first publication appeared at the beginning of January 1939; in a second publication at the beginning of February, we stated that thorium too is split by fast neutrons and we were able to demonstrate the second partner in the fission process to be the active rare gas, krypton. The sum of the atomic numbers 56 for barium and 36 for krypton makes up 92—uranium.

Immediately after our first publication on the chemical proof of barium isotopes obtained from uranium, Lise Meitner and Otto Robert Frisch explained this process, using Bohr's model of the atom, and made an estimate of the immense amount of energy released through the splitting, as shown by the mass-defect curve of the elements of the Periodic System. It was Meitner and Frisch who coined the word " fission."

A glance at the atomic weights of the relevant fission products, compared with the atomic weight of uranium, immediately showed us that the new artificial atoms must have much too high atomic weights, that they were therefore unstable and would be transformed by β-radiation into atoms of higher elements. Indeed it was through their activities that Strassmann and I had discovered the fission. We also spoke of the possibility that superfluous neutrons were evaporated in order to compensate for the too high atomic weights.

The proof of the emission of such additional neutrons during the fission process was discovered by Joliot, von Halban, and Kowarsky, as well as by American research workers. With this evidence that two or three (more accurately 2·56) additional neutrons are generated with the fission of uranium, chain reactions had become a possibility.

But even as early as 1939 it was recognized that the processes were much more complex than had originally been assumed. The fission only takes place in the case of the rare uranium isotope 235, while the much commoner uranium 238 is not split by the slow neutrons that we used. By emitting β-rays it is transformed into the next higher element, that is into a genuine transuranic element with the nuclear charge 93. This element, in its turn, again emitting β-rays, is transformed into the next higher element 94.

Dr. Meitner and I had found the beginning of these processes several years before in a β-ray-emitting artificial uranium isotope with a half-life period of 23 minutes; but with our weak radiation sources we had been unable to find the transformation products. Later these elements were discovered in the United States; element 93 was called neptunium and its successive product, element 94, became known as plutonium.

The atoms which were taken to be transuranic before the discovery of nuclear fission—first by Fermi and then by us—were all fission products, that is medium heavy elements.

In the year 1939 there was a very lively international and completely unrestricted discussion on the chemical and physical consequences of the newly discovered nuclear process. But in the autumn of 1939, with the outbreak of the second world war, silence reigned in the nuclear-physics institutes of the world.

The rest is a matter of general knowledge. The bombs on Hiroshima and Nagasaki showed what had been done in barely five years by the combined work of American, English, and Canadian physicists and chemists, helped by refugee scientists from Germany, Italy, and France. In Germany atomic physicists had also found that an energy-producing chain reaction was possible; it was however absolutely impossible to try to produce atom bombs during the war, and many scientists were glad not to have to shoulder such a gigantic responsibility.

We chemists in the Kaiser-Wilhelm Institute for Chemistry did not participate in the work of the physicists. We were interested in the clarification of the very complex fission processes and the isolation of the numerous isotopes. By the spring of 1945 we had set up a table of 25 different elements with more than 100 direct or indirect fission products of uranium. Even during the war we published all our results. The first comprehensive large-scale Table in the United States did not appear till after the war, nearly two years later. Strassmann and I felt a certain satisfaction when we found that the American Table included a number of half-lives merely designated as " short," which we, with our primitive means, had been able to pinpoint more precisely. The American radiation sources were of such intense strength that no such rapid work was possible.

We developed this isolation of short-lived transformation products almost as a sport. Thus, apart from a series of more stable alkali metals, we were able to isolate completely—that is, separate from the numerous other fission products—an active cæsium isotope with a half-life of about 40 seconds and an active strontium isotope with one of 80 seconds. In order to do this, we had recourse to the method I had introduced many years before, using radioactive rare gases, the so-called emanation method.

To obtain the short-lived alkali metals which arise in the fission from the still shorter-lived krypton and xenon isotopes, we used uranium preparations with a high emanation capacity, from which a large percentage of the active rare gases diffused at normal temperature despite their very short half-lives. On a negatively charged plate we collected the alkali metals as " active deposits." It was then possible to go to work so quickly on the invisible mixture of substances precipitated on to the plate that we were able, for instance, to register cæsium on our counter as cæsium silicotungstate 1·2—1·4 minutes after interrupting the radiation. By other methods, the time taken for the mixture cæsium–rubidium (separated from all other products) was 1·6—1·8 minutes.

When examining the very complicated mixtures of the rather longer-lived transformation products of the rare gases, we operated in the following way : During the irradiation of the uranium solution, the rare gases were passed through various freezing mixtures, one of which condensed the radioactive xenon while another condensed the krypton. In this way we had a direct method of separating the isotope group cæsium–barium–lanthanum from the group rubidium–strontium–yttrium.

I have come to the end of my survey. In August last year Dr. Strassmann and I attended the International Conference on the Peaceful Uses of Atomic Energy. We were extremely impressed by what we heard and what was shown to us there. In particular, we saw the quite extraordinary development attained in atomic nuclear research, not only in U.S.A., but also in England as well under the guidance of Sir John Cockcroft.

Wonderful indeed are the perspectives being opened up for the use of atomic energy in peace—may they blot out entirely the suicidal prospects of the use of atom and hydrogen bombs in war !