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OTTO HAHN

(Nobel Laureate for Chemistry, 1944)

NEW ATOMS

PROGRESS AND SOME MEMORIES

Few could have realised, when *Otto Hahn* split the uranium atom in 1938, that this was one of the momentous happenings of our time. Nor could anyone have expected that Hitler, at the full tide of power, would fail to turn to aggressive use the brilliant achievements of *Hahn* and his school. Yet we it was who made the atom bomb from an isotope of uranium, while *Hahn* held aloof from the Nazis, and even for long managed to protect his brilliant Jewish colleague *Lise Meitner*.

How did this gentle professor—in London not long ago at the invitation of the Government—make the discovery so potent for possible good and certain evil? Why did the Nazis make so little use of it? What are the elements we can now make, and what their potentialities for the future? Why is the hydrogen bomb so much more disastrous a prospect than even Bikini holds out?

Who else could say better than *Hahn* himself? In the three parts of this book we first hear his Nobel lecture on the artificial fission of uranium, then listen to an account of the new elements, including berkelium and californium, and finally enjoy some armchair reminiscences of the early days when *Hahn* collaborated with *Ramsay* in London, worked in Montreal at *Rutherford's* institute, and disputed with the American *Boltwood*; the days when apparatus often contrived from tin cans and cigarette boxes was opening up whole new worlds of science.

Intense personal interest always attaches to a great scientists' own account of his work: this book has topical interest too, because it makes very clear what many laymen want to know.

FROM REVIEWS OF THE GERMAN AND
DUTCH EDITIONS

'It is a particular attraction of this small publication to learn at first hand details of the by no means straight course of the history of the discoveries'. • *Experientia* (Switzerland)

'The present book may prove of considerable value to scientists who want a short, intelligent, and thorough account, without too much of the sugar and whipped cream of popularization'. *Acta Chemica Scandinavica*

'With a rare mastery *Hahn* knows how to introduce the reader almost painlessly to the most modern problems of nuclear physics, without assuming too much previous knowledge of the subject. In delightful contrast to other generally understandable books, the presentation leaves nothing to be desired as far as scientific rigorousness is concerned'.

Die Naturwissenschaften (Germany)

...expounded in a masterly fashion... a first-rate summary in understandable language... extremely interesting... no compilation but the work of a master... subtle humour...'

T.N.O.-Nieuws (Netherlands)

'We could not be better informed than by Prof. Otto Hahn himself, who has been involved in the story of radioactivity almost since the discovery of this phenomenon. This book is to be recommended warmly to those interested in this subject'.

Belgische Chemische Industrie (Belgium)

NEW ATOMS

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NEW ATOMS

PROGRESS AND SOME MEMORIES

by

OTTO HAHN

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A collection of papers, edited by

DR. W. GADE

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Let us hope that the tremendous possibilities connected with the utilisation of nuclear transformations will bring rich blessings to mankind and not destruction.
(OTTO HAHN)

PREFACE

Professor OTTO HAHN, who was awarded the Nobel Prize for Chemistry in 1944, is one of the world's outstanding authorities on radioactivity and atom research. At the end of 1938 he, together with his collaborator FRITZ STRASSMANN, discovered the fission of uranium and thorium. How this discovery was then worked out in detail and ultimately led to the utilisation of the energy liberated by nuclear fission may now be regarded as common knowledge.

At the period when the possibility of this practical utilisation was approaching realisation, HAHN's own country was under the sway of the Hitler regime, in whose hands atomic energy might well have spelt the doom of all mankind. Nothing could more eloquently testify to the humanitarian integrity of this scientist than the fact that, during the whole of World War II, he directed his work and that of his collaborators towards its utilisation for peaceful purposes only, though lack of equipment forced his hand to some extent. He maintained rigid silence on those other aspects which ultimately reached their sensational zenith in the American atom bomb. The writer never mentions the ensuing mental conflict in so many words, but it is sensed as ever-present between the lines of his writings. His personal acquaintances know, however, that that responsibility weighed heavily upon his shoulders.

This small volume unifies several of OTTO HAHN's publications into a single work revealing the author, as it were, in various surroundings. In Chapter I we meet him on one of the most impressive occasions known

to our society, which falls to the lot of only the most eminent torch-bearers of our culture, namely while delivering his Nobel lecture.

In Chapters II and III he talks to us, as from the platform in his lecture room, about nuclear physics generally, the potential uses for atomic energy and the new synthetic chemical elements in particular.

Finally, in Chapter IV, he introduces us, after terminating his lecture, into a less scientific atmosphere, dwelling pleasantly, and often with humour, on reminiscences of old-time researches into natural radioactivity, which he witnessed almost from the beginning and in which he took a leading part.

To make each chapter stand complete in itself, some passages have unavoidably had to be repeated.

Finally, I wish to thank those responsible for the English translation of the various parts of the book, especially Mr G. MIDDLETON, B. Sc., F.R.I.C. (London) for chapters I and II, Mr D. EVANS, B. Sc. (London) for chapters III and IV, Dr H. KRONBERGER (Harwell), and Mr H. W. LOEB, B. Sc. (London).

W. G.

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I. FROM THE NATURAL TRANSMUTATIONS OF URANIUM TO ITS ARTIFICIAL FISSION

The year 1946 marked a jubilee in the history of the chemical element, uranium. Fifty years earlier, in the spring of 1896, HENRI BECQUEREL had discovered the remarkable radiation phenomena of this element, which were at that time grouped together under the name of radioactivity.

For more than 100 years uranium, discovered by W. H. Klaproth in 1789, had had a quiet existence as a somewhat rare but not particularly interesting element. After its inclusion in the Periodic System by D. MENDELEJEFF and Lothar Meyer, it was distinguished from all the other elements in one particular respect: it occupied the highest place in the table of the elements. As yet, however, that did not have any particular significance.

We know to-day that it is just this position of uranium at the highest place of the then known chemical elements which gives it the important properties by which it is distinguished from all other elements.

The echo of BECQUEREL's fundamental observations on the radioactivity of uranium in scientific circles was at first fairly weak. Two years later, however, they acquired an exceptional importance when the CURIES succeeded in separating

from uranium minerals two active substances, polonium and radium, of which the latter appeared to be several million times stronger than the same weight of uranium.

It was only a few years before the first surprising property of this "radiating" substance was explained. The radioactive elements decompose according to definite rules into other active elements with different chemical and physical properties, simultaneously emitting corpuscular particles, the α - and β -rays. The α -rays were characterized by RUTHERFORD as positively-charged helium nuclei, after it had been shown earlier that the β -rays consisted of negative elementary particles, the so-called electrons.

With the hypothesis, suggested by RUTHERFORD and SODDY in 1902, of the disintegration of the atoms, the postulate of the indivisibility of chemical elements had to be abandoned.

Parallel with the investigation of the radioactive disintegration processes of uranium, which produce radium, radium emanation, the so-called active deposits, and polonium, research was carried out on thorium, at that time the second highest element of the Periodic Table. From thorium were obtained, in an exactly similar manner, strongly active disintegration products: radio-thorium, mesothorium, thorium X, etc. There was also a third series of disintegration products, the actinium series, which however also started from uranium.

By the systematic study of the penetration of α -particles

through thin layers of material, RUTHERFORD in 1911 was able to propose his model of the nuclei of chemical atoms. According to this view, the atoms of the elements consist of the positively-charged nucleus, representing practically the whole mass of the atom, around which move, at a relatively great distance, electrons which neutralize the nuclear charge. The charge of the nucleus undoubtedly represents the position of the element in the Periodic System.

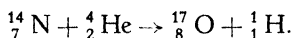
The radioactive disintegration reactions are thus nuclear changes which are not affected by the usual processes of physics or chemistry.

We are so certain of the unchangeability of these natural laws of radioactive breakdown, that from the speed of the changes we can construct a sort of "geological clock". Actually the final substance which is produced from uranium by way of these many active intermediate products is stable lead; from the quantity of the latter formed in the originally pure uranium mineral, it is possible to calculate the age of the mineral and therefore also of the geological deposit in which it has crystallised out. The same holds for thorium, which also produces as final product a kind of lead, although different from the "uranium lead."

Investigation proceeded further. Again the α -particles of radioactive substances formed the means for the solution of problems of nuclear physics. Following RUTHERFORD's nuclear model, these particles were recognized as free helium nuclei. With their relatively large mass of 4 (compared

with the small electron mass of about $\frac{1}{1800}$) and their initial velocity of up to 15,000 km/sec, they formed the bullets with which it might be possible to blast a way to the nuclei of the atoms, inaccessible to other means. And again it was the genius RUTHERFORD who made the next discovery, which was to have extremely important results. In 1919 he was able to show that by bombardment of nitrogen with the energetic α -particles of radium-C, an α -particle, that is a helium nucleus, was captured by the nitrogen nucleus, and a hydrogen nucleus—a proton—left the newly formed nucleus.

The process is represented by the equation:

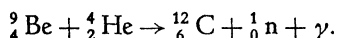


In this equation the lower indices represent the nuclear charges, the upper ones the nuclear masses (atomic weights). The remaining energy is given up in the form of kinetic energy to the released proton, which shows that the latter is derived from the nucleus.

Here for the first time was an artificial transmutation of atoms, actually a building-up of atoms, since an oxygen of mass 17 was produced from a nitrogen of mass 14.

In the following years a large number of reactions following the same pattern were discovered. On account of the positive charge of the α -particle this type of reaction did not succeed in bringing about nuclear transmutations of the heavy elements, since their high nuclear charge of positive sign repelled the α -particle sufficiently to prevent its penetration into the nucleus.

The year 1932 brought further important discoveries: the positron, heavy hydrogen, and—last but not least—the neutron. The discovery of the neutron was the result of detective work carried out in three countries. The initial impulse was given by BOTHE and BECKER in Germany, who were able to detect an exceptionally penetrating radiation, believed by them to consist of γ -rays, on exposing beryllium to α -particles. Then the JOLIOT-CURIES in France proved that the “ γ -rays” which appeared in the experiments of BOTHE and BECKER set free hydrogen nuclei (protons) with great energy from hydrogen-containing compounds, which is impossible for γ -rays. Finally CHADWICK in England gave the final explanation of these results: in addition to γ -rays there occurred uncharged neutral atomic nuclei with a mass of 1, which he called neutrons. The course of the reaction is:



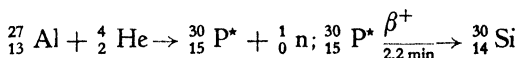
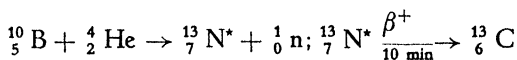
On the discovery of neutrons, the phenomenon of isotopy, known for a number of years, could readily be explained. The chemical elements are composed of protons and neutrons. The number of charged protons determines the chemical nature of the element: the sum of protons and neutrons determines the atomic weight. Thus a larger or smaller number of neutrons does not affect the chemical nature of the element, but merely produces isotopic kinds of atoms such as had been known for a long time in the different isotopes of lead, many mercury isotopes, two of chlorine, etc. Heavy hydrogen,

discovered in 1932, is similarly an isotope of ordinary hydrogen. It has in its nucleus both a proton and a neutron, and thus has a mass of 2 instead of 1. The highest chemical element known at that time, uranium, is also not a simple element, but contains isotopes of which U238 is the parent of the radium series and U235 that of the protactinium and actinium series.

Soon was it seen that neutrons would form projectiles specially suited for the transmutation of atoms: they have no charge and are thus not repelled by the positive nuclei of the elements. While however the (α, p) decompositions of RUTHERFORD— (α, p) is the abbreviated formula for a reaction in which an α -particle is taken up and a proton split off—nearly always produces stable kinds of atoms which occur naturally, in the (α, n) changes—the taking up of an α -particle and emission of a neutron—the JOLIOT-CURIES had observed an entirely new phenomenon. In 1934 they found that, on irradiation of certain elements with α -particles, not only neutrons but also positrons, that is positive electrons, appeared, even after the irradiation with α -particles had ceased. The emission of positrons fell off in the same way as had already been observed in the decay of natural radioactive elements. Artificial active elements had been produced. The first elements with which this “artificial” activity on irradiation had been observed were boron and aluminium.

From boron was produced an active nitrogen; from aluminium an active phosphorus; and in their turn these were

transformed into carbon and silicon respectively, with emission of positrons, according to the reactions



(* this indicates a radioactive isotope).

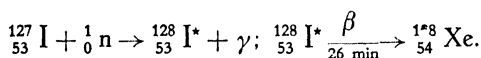
A new and large field for further investigation was hereby opened. At the same time the experimental possibilities for carrying out nuclear physical investigations were fundamentally expanded.

While so far the particles from natural radioactive elements had been the sole means of producing artificial nuclear reactions, high-voltage installations—van de Graaff generators and the cyclotron—could be now used in addition. More and more intensive irradiations could be carried out, and more and more new reactions could be realised.

For the time being, however, radium-beryllium mixtures continued to be used, since they were very convenient, though perhaps not very intensive, sources of neutrons for further investigations, especially of the newly-discovered artificial radioactivity. Such sources of neutrons were prepared by thorough mixing of a well-dried, finely-powdered radium salt with the finest possible beryllium powder, and sealing the mixture into airtight glass or metal tubes. Uranium itself was only of importance because radium was obtained from it.

It was especially the Italian scientist FERMI who first realised the great importance of neutrons for the production of nuclear reactions: FERMI and his co-workers irradiated practically all of the elements of the Periodic System with neutrons, and made numerous artificial radioactive elements.

Generally, the course of the reaction was such that, especially when working with slow neutrons¹, the neutrons were captured by the nucleus. Thereby in many cases an unstable isotope of the irradiated element was produced, passing into the next higher element with emission of β -rays. This latter was generally a stable atom occurring among the usual chemical elements, e.g.



FERMI and his co-workers continued their tests through the whole of the Periodic System up to uranium. Here also they discovered many transmutations produced by neutrons, including some very rapid ones. They proceeded from the obvious assumption that initially there are produced artificial, active, short-living uranium isotopes; as these emit β -rays FERMI inferred the production of so-called "transuraniums", representatives of the element 93 which is not known naturally, and possibly even of the still higher element 94.

FERMI's proofs were not accepted everywhere. It was pointed

¹ As FERMI first showed, such slow neutrons, of low energy, can be obtained from the primary high-energy neutrons by passing them through substances with a high content of hydrogen. The kinetic energy of the neutrons is then transferred to the hydrogen nuclei in elastic collisions.

out that for example in the case of the so-called 13-minute element—that detected with the greatest certainty—the possibility of its being an isotope of element 91, i.e. protactinium, could not be ruled out¹.

At this point LISE MEITNER and I decided to repeat FERMI's experiments in order to decide whether the 13-minute element was a protactinium isotope or not. This decision was taken the more readily since, by the discovery of protactinium (1917), we were familiar with its chemical properties. Moreover, a β -radiating isotope of element 91 was well known to us in the form of Uranium Z, discovered by myself, which had the favourable half-life of 6.7 hours, and was available from uranium salts.

With the help of the "indicator method" we were able to prove without doubt that the 13-minute element of FERMI was neither a protactinium isotope, nor a uranium, actinium or thorium. In accordance with the position of science at the time, FERMI's assertion should be correct, and the 13-minute element a representative of the element 93, that is a "transuranium".

We should point out here that other possibilities did not occur to anyone at that time. Since the discovery of the neutron and the application of artificial sources of radiation, a large number of most unusual nuclear reactions had been discovered; the products were always either isotopes of the

¹ From another direction (IDA NODDACK) the objection was raised that all the elements of the Periodic System must first be excluded before it was possible to draw the conclusion that an element 93 had been obtained. This objection was not taken seriously, as it appeared to be in opposition to all physical views of nuclear physics.

irradiated substances, or their next, or at most next-but-one, neighbours in the Periodic System; the possibility of a breakdown of heavy atomic nuclei into various light ones was considered as completely excluded.

With the tests on FERMI'S 13-minute element and the checking of other, rather less certain, results of FERMI, we found (later in co-operation with F. STRASSMANN) that the phenomena associated with the irradiation of the highest element of the Periodic System were much more complicated than had originally been supposed. FERMI and his co-workers had already, in their first communication, described two short-life β -radiating kinds of atoms (half-life 10 sec and 40 sec), which they naturally considered to be artificial isotopes of uranium produced from the original uranium by the capture of neutrons. LISE MEITNER and I found, in addition, a substance with a half-life of 23 minutes, which we conclusively identified as an artificial radioactive uranium isotope. With FERMI'S substances of short life, the isotopy with uranium can only be assumed, but not proved. The 23-minute element occurred without any other radiation conditions in a so-called "resonance process".

As the result of many years of work, we (HAHN, MEITNER and STRASSMANN) had finally obtained a great number of artificial active kinds of atoms, which all appeared to be formed directly or indirectly by β -radiation from the supposed short-living uranium isotopes, and which therefore must all represent so-called transuraniums—elements higher than uranium.

According to their chemical behaviour, these could be classified into various groups, and, since in many cases the gradual production from β -radiating parent substances could be directly observed, decay schemes were drawn up extending to elements 95 and 96. In so far as the work was repeated by others, the results were always confirmed.

Independently of the transuranium investigations of HAHN, MEITNER and STRASSMANN just mentioned, CURIE and SAVITCH described in 1937 and 1938 a so-called 3.5-hour substance which they had obtained by irradiation of uranium with neutrons, and of which the chemical properties could not readily be determined. According to CURIE and SAVITCH, the substance appeared to be a rare earth, but was not actinium; it had more resemblance to lanthanum, and could only be separated from the latter by "fractional crystallization". With some hesitation CURIE and SAVITCH decided to include the substance in the transuranium series, but the possibilities put forward by them appeared difficult to understand and unsatisfactory.

As this 3.5-hour element had been included with the transuraniums, I, together with STRASSMANN, tried to obtain it. After careful experiments we arrived at remarkable results, which may be formulated approximately as follows: "In addition to the transuraniums described by HAHN, MEITNER and STRASSMANN, there are produced by two successive α -emissions three artificial, β -active radium isotopes with different half-life times, which in their turn change into artificial β -active

actinium isotopes". The conclusion that radium isotopes had been produced was the only one possible since, according to the chemical properties, only barium and radium could be considered. Barium was, according to the physical viewpoint of the time, impossible, and thus only radium was left.

The separation of this active group was performed by means of a barium precipitate; not however in the form of barium sulphate, which with its large surface strongly adsorbs other elements, but, on the suggestion of STRASSMANN, as barium chloride, which crystallizes very well from concentrated hydrochloric acid and which precipitates uncontaminated by other substances.

At the same time the production of radium under these conditions of radiation was very remarkable: α -decompositions had never been observed with neutrons low in energy, and yet here, as with the transuraniums, a number of isotopes appeared simultaneously.

The experiments were continued in various directions. The preparations were, however, always very weak and the α -rays of the most stable of the new isotopes were so strongly absorbed that thicker layers could only be investigated with poor yields of radiation. An attempt was therefore made to separate the artificial "radium" as far as possible from the barium added as carrier, in order to obtain coatings permitting easier measurement. This was done by fractional crystallization using the method of Madame CURIE, a method with which we had been thoroughly familiar over a number

of years. About 30 years previously I, together with **EISE MEITNER**, had separated the radium isotope mesothorium from barium by fractional crystallization. More recently, with the assistance of a number of co-workers, the laws governing the formation of mixed crystals between radium and barium salts had been systematically investigated.

The attempts to separate our artificial "radium isotopes" from barium in this way were unsuccessful; no enrichment of the "radium" was obtained. It was natural to ascribe this lack of success to the exceptionally low intensity of our preparations. It was always a question of merely a few thousands of atoms, which could only be detected as individual particles by the **GEIGER-MÜLLER** counter. Such a small number of atoms could be carried away by the great excess of inactive barium without any increase or decrease being perceptible, even if the barium was precipitated in the form of barium chloride, which precipitates in a very pure form.

In order to check this, we repeated the same tests with a weak intensity of the natural radium isotopes mesothorium and thorium X. These substances were freed from every trace of their parent substance and decay products with the greatest care and, by systematic dilution, preparations were made which were only just detectable with the **GEIGER-MÜLLER** counter. Crystallizations were carried out with the chlorides, bromides and chromates, always with the corresponding barium salt as carrier.

The result was, as was to be expected for radium, that

mesothorium and thorium X were concentrated in the first fractions of the salts named, and in fact in quantities such as we should expect from our previous experience. This proved that the few atoms of natural radium isotopes also behaved in exactly the same manner as strong preparations.

Finally we proceeded to direct "indicator tests". We mixed the pure natural radium isotopes with our artificial "radium" isotopes, also previously freed from their decay products, and fractionated the mixture in the same way as before. The result was that the natural radium isotopes could be separated from barium, but the artificial ones could not.

We checked the results in still another way. If the artificial alkaline earth isotopes were radium, then the decay products produced directly through β -emission should consist of actinium: from the element 88 should be produced the element 89. If on the other hand it was barium, then lanthanum should be formed: from element 56 the next higher element 57. With the aid of the pure actinium isotope mesothorium-2 we carried out an "indicator test" by mixing mesothorium-2 with one of the known primary decay products of artificial radium isotopes, and then carrying out the chemical separation of actinium and lanthanum by the method of Madame CURIE. During the fractionation of lanthanum oxalate with actinium, the latter accumulates in the final fractions. This actually occurred with the actinium isotope mesothorium-2. The decay product of our so-called "radium isotope" however remained with the lanthanum. The artificial rare earth, which had been

considered to be actinium, was really lanthanum. Thus it was established that the alkaline earth isotope, which we had believed to be radium, was in fact an artificial active barium; the lanthanum could have been produced only from barium and not from radium.

In order to make quite certain, we carried out a so-called "cycle" with barium. The most stable of the active isotopes, now identified as barium, was freed from active decay products and other impurities by recrystallization with inactive barium; one quarter of the total quantity was kept for comparison, and three quarters were subjected to the following cycle of barium precipitations: Ba chloride $\rightarrow$ Ba-succinate $\rightarrow$ Ba-nitrate $\rightarrow$ Ba-carbonate $\rightarrow$ Ba-ferrimannite $\rightarrow$ Ba-chloride. After passing through this series of compounds, many of which crystallized beautifully, the resulting barium chloride and the recrystallized comparison preparation were measured alternately using the same counter, with equal weights and equal thicknesses of layers. The initial activity and the increase as the result of further formation of the active lanthanum were the same for both preparations, within the limits of error: the crystallization of so many and such different salts had produced no separation of the active barium from the carrier. It could only be concluded that the active product and the carrier were chemically identical, that is, barium.

In the first communication on these tests, which "were in opposition to all the phenomena observed up to the present in nuclear physics" (January 6th, 1939), the indicator tests

mentioned had not been entirely completed, and we had therefore expressed ourselves cautiously. As a second partner in the new process we assumed an element with an atomic weight of about 100, as in that case the combined atomic weights would be that of uranium, "for example $138 + 101$ (e.g. element 43) gives 239!"

After the completion of the measurements in hand, and of the "cycle", the possibility of error was still further excluded.

This completion of the tests and the above-mentioned "cycle" appeared in a second communication (February 10th, 1939). This also described the splitting of the element thorium and its confirmation with the aid of indicator tests analogous to those described above. Here also reference was made to the detection of an inert gas and an alkali metal derived from it; the nature of the gas was recognized, and its separation from uranium accomplished, by means of a current of air passed over the uranium during the irradiation. An active strontium and an active yttrium were identified in the uranium itself.

Immediately after the first publication on the production of barium from uranium, there appeared as a first communication an article by LISE MEITNER and O. R. FRISCH in which the possibility of a breakdown of heavy atomic nuclei into two lighter ones, with total charges equal to that of the original nucleus, was explained with the aid of BOHR's model of the original nucleus. MEITNER and FRISCH also estimated the exceptionally high energy output to be expected from this

reaction, from the curve of the mass deficiencies of the elements in the Periodic Table. The great repulsive energy of the fragments produced by the splitting was first demonstrated experimentally by FRISCH and shortly afterwards by F. JOLIOT. MEITNER and FRISCH soon proved that the active breakdown products, previously considered to be transuraniums, were in fact not transuraniums but fragments produced by splitting. They were able to accumulate these by "repulsion" outside the radiated uranium.

In quick succession there appeared a whole series of publications from European and American nuclear physics institutes, confirming and expanding the tests described.

Thus the process proceeds in such a way that the nucleus of the uranium with a charge of 92 is split into two nuclei of moderate size¹. If one of these is barium, which has a nuclear charge of 56, there must be produced at the same time a krypton with a nuclear charge of 36. Together these nuclei add up to 92. Both have however, as may easily be seen from the masses of uranium and of the stable isotopes of barium and krypton which occur naturally, too great a mass, and thus an excess of neutrons. They should therefore pass over into stable elements with higher nuclear charges, with emission of β -rays; and in fact, as our later experiments showed, sometimes achieve stability by way of a great number of unstable intermediate decay products.

¹ The expression "nuclear fission" (Kernspaltung, fission nucléaire) is due to MEITNER and FRISCH.

The highest stable krypton isotope has a mass of 86. In uranium fission there is produced, among other atoms, an unstable krypton with mass 88. Uranium 235 is responsible for the fission induced by thermal neutrons, as BOHR was the first to see; this fission forms by far the larger part. If there are no side reactions then the mass of the other fission product belonging to the krypton 88, that is of the barium, should be $236 - 88 = 148$. As the highest stable barium isotope has a mass of 138, the first-mentioned product is not less than 10 units heavier. STRASSMANN and myself had already noted, in our second communication, the possibility that neutrons were set free in the fission process. That this was in fact the case was first established experimentally by F. JOLIOT.

The investigations continued at a rapid pace, both from the physical and the chemical side. Only a year after the first communication on the production of barium from uranium, there appeared in the *Reviews of Modern Physics* (U.S.A.) a bibliography on the splitting of heavy nuclei (Nuclear Fission, by L. A. TURNER) in which nearly one hundred publications in this sphere were mentioned.

During the second world war, the very confusing fission reactions were systematically investigated in the Kaiser Wilhelm Institute for Chemistry with a view to their chemical disentanglement, and numerous new reactions were discovered. Japanese investigators found that, when fast neutrons were used, the fission of uranium proceeded more symmetrically than with slow ones. At the beginning of 1945 we were able

Table 1

Atomic weights of some natural isotopes, and of artificial isotopes produced by nuclear fission, if the excess of neutrons is not released.

Element	Natural isotopes	Isotopes occurring in the uranium fission
54 Xenon	124, 126, 128, 129, 130, 131, 132, 133, 134, 136	139 (unstable) together with
38 Strontium	84, 85, 87, 88	97 (unstable)
—		—
92	.	236
36 Krypton	78, 80, 82, 83, 84, 86	88 (unstable) together with
56 Barium	130, 132, 134, 135, 136, 137, 138	148 (unstable)
—		—
92		236
92 = nuclear charge of uranium = sum of the nuclear charges of the fragments		236 = mass number of uranium 235 plus 1 neutron

to make a table in which were collected, as direct or indirect products of uranium fission, 25 different elements, ranging from 35 (bromine) to 59 (praseodymium), in the form of about 100 active kinds of atoms. A larger and more extended list was published in America at the end of 1946, as the total result of research in the U.S.A. into the course of the fission process. This summary, reproduced here in Table 2 (p. 30–33) begins at element 30 (zinc) and goes up to element 64 (gadolinium). The yield of those fission products which were not mentioned by us in 1945 is however so small that we could not have detected them with the ineffective sources of radiation available to us (compared with the very much stronger ones of America).

The active atoms, believed by us up to 1939 to be trans-uraniums, were all fission products and their active successors, and not elements with atomic number higher than uranium.

From the nature of the problem, the physical work proceeded in a different direction. Especially important in this connection was the above-mentioned investigation of JOLIOT in which he proved experimentally, in the spring of 1939, that in the fission process neutrons appeared in addition to the (always two) new elements.

Since by the action of neutrons on uranium, fresh neutrons are liberated, the latter, if they meet uranium atoms, produce further fissions in their turn. If more than one fresh neutron is produced, and the process is so arranged that all the fresh neutrons strike uranium atoms, then we have a chain of

continuously-renewing fission reactions which, like an avalanche started by a snowball, can attain enormous dimensions. Thereby the practical application of atomic energy first came into the range of possibility. S. FLÜGGE, then attached to the Kaiser Wilhelm Institute for Chemistry, was the first to refer to this.

About 10 years ago, JOLIOT concluded his Nobel lecture with the following words: "If we look back and take a glance at the progress achieved in ever-increasing measure in science, then we are justified in supposing that the investigators who are able to build up or break down elements at choice, will also learn how to realise transformations of an explosive character; veritable chemical chain reactions."

"If it should happen that such transmutations are propagated in matter, then one can easily imagine the enormous liberation of free energy which will take place. But unfortunately, if the infection is carried to all the elements of our planet, we must anticipate with apprehension the consequences of the release of such a cataclysm. From time to time astronomers have observed that a star of moderate magnitude suddenly increases in size; a star invisible to the naked eye may become very brilliant and visible without instruments: this is the production of a *nova*. Such a sudden illumination of a star is possibly caused by transmutations of an explosive character like those which our wandering imagination is perceiving now — a process that the investigators will no doubt attempt to realise while taking, we hope, the necessary precautions."

Table 2. Summary of the fission products of uranium known at the end of 1946 (U.S.A.) Mass numbers and half-life times in brackets are uncertain; dotted arrows indicate uncertain transformations.

s = seconds; m = minutes; h = hours; d = days; a = years.

Mass	Atom No.	30	31	32	33	34	35	36	37	38	39	40	Fission yield %
		Zn	Ga	Ge	As	Se	Br	Kr	Rb	Sr	Y	Zr	
71			stable										
72		4.9h →	14.3h →	stable									$15 \cdot 10^{-3}$
73		< 2m →	5h →	stable									$10 \cdot 10^{-4}$
74				stable									
75				(69m) →	stable								
76				stable									
77				12h →	40h →	stable							
78				(80m) →	stable								
(78)				2.1h →	90m →	?							
79					< 10m or > 7 · 10 ⁴ a	→ stable							0.0091
80					stable								0.02
81					59m →								
82					17m →	stable							0.008
83					34h →	stable							0.125
84					25m →	24h →	113m →						$2.8 \cdot 10^{-5}$
85					~ 2m →	30m →	stable						0.40
86					3.0m →	4.5h →	stable						0.65
(87)					55.6s →	stable	19.5d →	stable					~ 0.24
87					50s →	75m →	6.3 · 10 ⁴ a →	stable					$2 \cdot 10^{-3}$
88					3h →	17.8m →	stable						0.026
89					2.6m →	15.4m →	5.3d →	stable					4.6
90					~ 33s →	short →	25a →	65h →	stable				
91								51m →	51m (~ 40%)				
92								9.8s →	97h →	57d →	stable		5.9
....								3s →	short →	2.7h →	3.5h →	stable	5.1
								80s →	?				

Atom No.	36	37	38	39	40	41	42	43	44	45	46	47	48	49	50	Fission yield %
Mass No.	Kr	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	
(94)	20s→short→1.4s→short→1m→20m→stable	short→short→1m→20m→stable	1m→20m→stable	10h→20m→stable	?	stable										~5
95		o	o	o	90h (~2%)											~6.4
96		o	o	o	65d→35d→stable											
97	short→short→11.0h→75m→stable	short→short→11.0h→75m→stable	short→short→11.0h→75m→stable	stable												
98																
99																
100																6.2
101																
(102)																
103																37
104																~0.9
105																0.5
106																
107																
108																
109																0.028
110																0.018
111																0.011
112																
113																
114																
115																0.011
116																0.0008
117																0.01
...																

Table 2 (continued)

Mass	Atom No.	50	51	52	53	54	55	56	57	58	59	60	Fission yield %
118		stable	•	•	•	•	•	•	•	•	•	•	
119		stable	•	•	•	•	•	•	•	•	•	•	
120		stable	•	•	•	•	•	•	•	•	•	•	
(121)		62h → stable	•	•	•	•	•	•	•	•	•	•	0.014
(121, 123)		130d → stable	•	•	•	•	•	•	•	•	•	•	0.0012
122		stable	•	•	•	•	•	•	•	•	•	•	
(123)		70d → stable	•	•	•	•	•	•	•	•	•	•	0.0044
124		stable	•	•	•	•	•	•	•	•	•	•	
(125)		(9m) → ~27a → stable	•	•	•	•	•	•	•	•	•	•	0.023
(125)		~20m → ?	•	•	•	•	•	•	•	•	•	•	
(126)		70m → 60m → stable	•	•	•	•	•	•	•	•	•	•	0.1
127		•	•	90d	•	•	•	•	•	•	•	•	0.033
128		•	93h → 93h → stable	•	•	•	•	•	•	•	•	•	
129		•	•	stable	•	•	•	•	•	•	•	•	0.19
130		•	4.2h → 70m → v. long → stable	•	•	•	•	•	•	•	•	•	
131		•	•	30h	•	•	•	•	•	•	•	•	~0.5
(132)		•	•	25m → 80d → stable	•	•	•	•	•	•	•	•	2.8
133		•	~5m → 77h → 24h → stable	•	•	•	•	•	•	•	•	•	3.6
(134)		•	<10m → 60m → 22h → 5.3d → stable	•	•	•	•	•	•	•	•	•	~4.5
••••		•	<10m → 43m → 54m → stable	•	•	•	•	•	•	•	•	•	~5.7
135		•	•	~1m → ?	•	•	•	•	•	•	•	•	
		•	•	•	13m (~10%)	•	•	•	•	•	•	•	
		•	•	<2m → 6.7h → 9.2h → 2.5·10 ⁶ → stable	•	•	•	•	•	•	•	•	

Mass	Atom No.	53	54	55	56	57	58	59	60	61	62	63	64	Fission Yield %
(136)	I	18m→stable←13d→stable												0.07
(137)	Xe	22.0s→?												0.17
137		30s→34m→33a→stable												
138		17m→32m→stable												
139		41s→7m→85m→stable												6.3
140		16s→short→12.3d→40.0h→stable												6.1
....		40s→?												
141		3s→short→18m→37h→28d→stable												5.7
(142)		~1-2m→6m→74m→stable												
143		1s→short→<0.5m→19m→33h→13.8d→stable												5.4
144		short→short→short→275d→17.5m→stable												5.3
(145)		0.8s→short→short→short→1.8h→4.5h→stable												
....		(68m)												
(146)							14.6m→24.6m→stable							2.6
147							11.0d→3.7a→stable							
148							stable							1.4
149							(17h)→47h→stable							
150							stable							
(151)							(short)→12m→long→stable							
152							stable							
153							<5m→47h→stable							0.15
154							stable							
155							25m→2a→stable							~0.03
156							<5m→~10h→15.4d→stable							0.073
157							15.4h→stable							0.0074
(158)							60m→stable							0.002

What was ten years ago only a figment of our "wandering imagination", has already become to some extent a threatening reality. The energy of nuclear physical reactions has been given into men's hands. Shall it be used for the assistance of free scientific thought, for social improvement and the betterment of the living conditions of mankind? Or will it be misused to destroy what mankind has built up in thousands of years? The answer must be given without hesitation, and undoubtedly the scientists of the world will strive towards the first alternative.

Postscript

In the preceding paragraphs we have given a general outline of how the investigation of the natural radioactivity of uranium led to the artificial splitting of that element. But this does not exhaust the possibilities of uranium. Uranium consists mainly of two isotopes with atomic weights 235 and 238. The first is present in the mixture only to the extent of $1/140$ part; nevertheless the fission process described above, which is caused with special violence by "slow" neutrons, is to be ascribed chiefly to its rare isotope. It has already been mentioned above that LISE MEITNER and the present writer were able to identify, after the irradiation of uranium with neutrons, a substance having a half-life of 23 minutes as undoubtedly a uranium isotope. (The short-lived isotopes, thought first by FERMI and later by ourselves to be artificial uranium isotopes, were actually fission products.) The 23-minute

uranium was produced in a so-called resonance process by neutrons of a certain velocity. Since this substance emits β -rays, a representative of element 93, that is an actual transuranium, *must* be produced. Although we sought for it, we were unable to detect it in the very weak preparations which we had at the time. Later it was identified in the United States as a β -emitter with a half-life of 2.3 days. Thus it has an atomic weight of 239. Since this true transuranium 93 emits β -rays in turn, it must be concluded that an element 94, a further transuranium, is formed. This is a relatively long-living α -emitting substance (half-life 24,000 years). The American scientist SEABORG gave it the name plutonium, calling element 93 neptunium. A second isotope of element 93 also occurs, it is produced in another way from uranium 238, namely by a so-called (n, 2n) process with the aid of neutrons of great energy: one neutron, so to speak, in passing through the uranium nucleus knocks out another one. The result is a β -emitting uranium isotope of mass 237, which thus transforms into a second neptunium isotope of which the half-life is some millions of years.

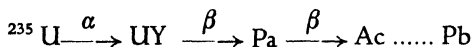
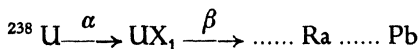
Thus the behaviour of uranium on irradiation with neutrons of different velocities, both fast and slow, is very complicated (see Table 3). In addition to the natural splitting process, which continues during the irradiation at a speed independent of all the other reactions, the following occur:-

(1) Nuclear fission with formation of numerous artificial atoms of all elements between 30 and 64.

Table 3.

URANIUM TRANSMUTATIONS

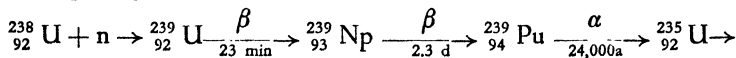
I. NATURAL



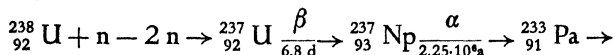
II. ARTIFICIAL

(i) *Fission processes* with slow and fast neutrons, up to the present 34 elements in about 200 kinds of atoms.

(ii) *Capture of a neutron:*



(iii) *Loss of a neutron:*



(2) Emission of surplus neutrons during this fission process, making a chain reaction possible.

(3) The resonance capture of a neutron with a definite energy by uranium 238, with formation of uranium 239, which in its turn is transformed into the elements neptunium and plutonium.

(4) The giving up of a surplus neutron by the U238 with formation of a U237, which also forms a neptunium isotope.

As, in process (1), slow (thermal) neutrons mainly attack the rare isotope U235, process (3) is a kind of competitor of process (1), since in the resonance process (3) the neutrons are captured before they have reached thermal velocities. It

is a matter of experimental technique to prevent this capture as much as possible, in order to facilitate the chain reaction. On the other hand, the extra neutrons produced in the fission of process (2) give rise to the resonance process, and thereby to the formation of plutonium. If this latter element is obtained in sufficient quantity by means of a slow controlled chain reaction carried out on a large scale (in a so-called "pile"), and is separated from uranium, then it can also act as carrier of a chain reaction. The same holds for pure U235 after it has been separated from the isotope 238, since with U235 the resonance process, which interrupts the chain reaction, does not occur.

Both uranium 235 and plutonium are made in the United States. The result was the bombing of Hiroshima and Nagasaki.

II. THE CHAIN REACTION OF URANIUM

1. THE ATOMIC NUCLEUS AND ITS DECOMPOSITIONS

The atomic nucleus of every chemical element consists of protons and neutrons. *Protons* are the positively-charged nuclei of hydrogen atoms: *neutrons* are uncharged atomic nuclei with practically the same mass as the protons. As "naked" atomic nuclei they are not surrounded by an electron cloud, and therefore form no chemical compounds. They merely play a part—though a very important one—in the building-up of the atomic nuclei and in nuclear reactions.

The number of protons, that is, of charged nuclear components, determines the chemical nature of the element, while the mass—the atomic weight—is determined by the sum of protons and neutrons. Since the neutron has no charge, a larger or smaller number of neutrons does not alter the chemical properties of the element but merely forms "isotopic" kinds of atoms with different masses which all belong to the one element characterized by the positive nuclear charge. These isotopes determine the "practical atomic weight" of that element.

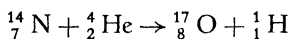
Practically all elements, from hydrogen—which consists of ordinary hydrogen of mass 1 with approximately one seven-

thousandth part of "heavy" hydrogen of mass 2—to uranium, are mixtures of isotopes; so-called *mixed elements*, in opposition to *simple elements* which consist of one kind of atom only.

Natural uranium consists of three isotopes, of which only the two chief ones interest us here; that is, the one of mass 238 and the rare one of mass 235.

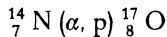
The first *artificial transmutation* of one element into another was due to RUTHERFORD, who in 1919 converted nitrogen into oxygen by bombarding it with α -particles, one proton being ejected. This reaction was detected by the greater range of this hydrogen nucleus in comparison with that of the α -rays.

Chemically this process may be written as follows:



(nuclear charge below left, mass above left, before the symbol of the element)

In physics it may be written more briefly as:



(α -particle = helium nucleus, p = proton = hydrogen nucleus)

The first letter between the brackets always represents the particle taken up, the second letter the particle ejected.

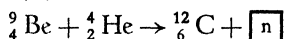
This method of writing a nuclear reaction, in general use to-day, will be dealt with further below. Radioactive decay reactions are shown by giving the half-life of the substance formed and the radiation given off.

In the beginning of the thirties further important discoveries were made: in 1932 that of the *neutron* by the English physicist CHADWICK, based on the investigations of BOTHE

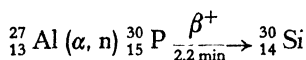
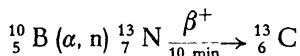
and BECKER in Germany and of the JOLIOT-CURIES in France. As has been stated above, the neutron, like the proton, has a mass of 1, but has a nuclear charge of 0. Thus it has no negative electron to neutralize a positive nuclear charge.

The discovery of the neutron (n) resulted from irradiation of beryllium with α -particles according to the reaction

${}^9_4\text{Be} (\alpha, n) {}^{12}_6\text{C}$, or described chemically:



In 1934 there followed the discovery of *artificial radioactivity* by the JOLIOT-CURIE's. They irradiated boron and aluminium with α -particles and obtained an unstable, that is radioactive, nitrogen and an unstable radioactive phosphorus. A neutron was also set free. The reactions were as follows:



Both the unstable atoms decayed further with emission of a β -particle = positive electron. The nuclear charge of the element formed is thus one unit lower than that of the original element.

In actual atomic transmutations, or rather *nuclear transmutations*, the number of protons and neutrons which form the nucleus is altered. A change in the number of protons signifies an actual change of *element*, the number of charged nuclear constituents being always altered. A change in the number of neutrons signifies, on the other hand, the production

of a different *isotope* of the same element, for there is no change in the nuclear charge, only in the atomic weight.

After the discovery of neutrons and of artificial radioactivity, it was soon seen that neutrons were a specially favourable means for bringing about transmutations of atoms, as they have no charge and therefore are not repelled by the positively charged atomic nuclei.

The mechanism of such a nuclear transmutation is, in most cases, that the neutron, when it approaches the nucleus, is taken up by the latter forming an isotope of the atom in question with a mass increased by the mass of the neutron, i.e. by 1. These artificial isotopes are however in most cases unstable. They do not occur naturally in the ordinary mixed elements, and pass over into the next higher elements with emission of β -rays, that is, of negative electrons.

2. THE FISSION OF URANIUM

The Italian physicist FERMI, the first to use neutrons for nuclear reactions, continued his experiments up to uranium, with the result that here also he discovered artificial transmutations with emission of β -rays. According to the state, at that time, of our knowledge of the binding power of atomic nuclei with high charges, these changes must lead to elements with atomic weights higher than uranium. Together with MEITNER, I was able to confirm and extend these experiments; then, with F. STRASSMANN in the years 1935-1938, a whole series

of artificial radioactive kinds of atoms, believed to be trans-uraniums, were discovered.

Our results led us to the conclusion that we were dealing with radioactive representatives of the elements 93, 94, 95 and 96. The proof of this was seen in the fact that these substances, according to their chemical properties, were certainly not isotopes of the left-hand neighbours of uranium in the Periodic Table, for example of protactinium or thorium. On the other hand the numerous kinds of atoms obtained belonged to at least four different elements.

Physicists considered it to be quite impossible that an atomic nucleus with a large charge like uranium, with its strong binding power resulting from the 92 positive nuclear charges, should be brought to complete disintegration by irradiation with neutrons. In hundreds of artificially-induced nuclear reactions with nearly all of the elements of the Periodic System, the reactions observed had always been ones in which the reaction products were either isotopes or the nearest neighbours of the irradiated elements. In so far as our experiments were repeated in other countries, they were all confirmed.

In the autumn of 1938, STRASSMANN and myself found, however, in connection with experiments of CURIE and SAVITCH, some very surprising results. We separated three alkaline earth metals, which we originally regarded merely as artificial radium isotopes. This was in itself remarkable, since radium, with its nuclear charge of 88, is not a next neighbour of uranium. Actually the results were even more puzzling.

Our “radium isotopes” could not be separated from barium by any of the methods for the separation of radium and barium with which we were familiar.

The problem was finally solved by indicator tests, in which we mixed our artificial “radium isotopes” with natural radium isotopes and then carried out the separation. The “natural” radium could be separated from barium, the artificial isotopes could not. On the basis of this observation we were forced to conclude that the alkaline earth isotopes, produced by the action of neutrons on uranium, were in fact not radium at all, but barium – a highly unexpected result. *By the bombardment with neutrons the uranium had been split into two nuclei of moderate weight.* The one was barium, the other, as was soon discovered, krypton. Their nuclear charges, 56 and 36 respectively, add up to 92 (cf. pp. 25 and 27).

MEITNER and FRISCH were the first to give the explanation of this process, using BOHR's atomic model. To them¹ also, as mentioned in chapter I, is due the expression *nuclear fission* (Kernspaltung, fission nucléaire).

The subject developed rapidly. It is described in detail in the technical literature of 1939.

As already stated on p. 34, the rare, lighter, uranium isotope with mass 235, which occurs in the mixed element uranium only to the extent of 1 part in 140, is of greatest importance in the fission, which is brought about especially by those neutrons which have lost their original high kinetic energy

¹ L. MEITNER and O. R. FRISCH, *Nature*, 143 (1939) 239.

by impact with other atoms, so that their velocity is comparable with that of ordinary gas molecules (thermal neutrons).

It was soon noticed that the splitting process could proceed in several ways: the fragments produced are not always isotopes of barium and krypton, but include a number of others, such as strontium and xenon, bromine and lanthanum, iodine and yttrium, etc., etc. The sum of the nuclear charges of the two fission products is always 92.

In 1944, at the Kaiser Wilhelm Institute for Chemistry, we were able to produce a table of fission products which included all elements from bromine (35) to praseodymium (59), mostly in the form of a number of isotopes with different half-lives. Sometimes these passed by way of a number of active intermediate products, and with emission of β -radiation, into the stable, and no longer detectable, isotopes of the next higher elements.¹

At the end of 1946 there appeared in the United States a very extensive report² on the work done there during the second world war. In this the series of fission products begins at zinc (30) and ends at gadolinium (64). These lightest and heaviest fragments occur however in such small amounts that—as already stated on p. 28—they could not have been detected with the sources of radiation available to us.

The substances which, before the discovery of the uranium

¹ See e.g. O. HAHN, *Veröffentlichungen des Deutschen Wissenschaftlichen Instituts Stockholm* No. 1 (1944); more completely in W. SEELMANN-EGGEBERT and F. STRASSMANN, *Z. Naturforsch.*, 29a (1947) 80.

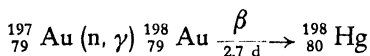
² "Plutonium Project", *J. Am. Chem. Soc.*, 68 (1946) 2411; cf. table 2, pp. 30–33.

fission, were considered by FERMI, by ourselves, and by other nuclear physicists to be transuraniums, were in reality all fission products of uranium, and thus active representatives of elements of moderate atomic weight. The actual transuraniums, which will be mentioned in more detail later, were present at that time in such small quantity that they were not detected.

In addition to the work on the elements which appear in the fission of uranium, there were further investigations of the normal nuclear reactions of the other elements, that is, of the reactions whereby active isotopes of the irradiated element or of its next neighbour are produced. This was done especially in America, with the aid of the cyclotron, of which there were a number in the United States, although there were none in Germany. Thus there was a possibility of obtaining active isotopes of nearly all the chemical elements, including ones previously unknown.

What are the *actual weights* which are obtained in these reactions? Even with the use of the most powerful high-voltage installations for the production of high-energy particles such as protons, deuterons (nuclei of heavy hydrogen) and neutrons, not to mention the neutrons from radium-beryllium mixtures, the artificial active isotopes obtained with the aid of these rays were far below the limits of normal chemical detectability or weighability. Even the great cyclotron of LAWRENCE in the University of California at Berkeley was, until a few years ago, unable to produce weighable quantities.

The detection and further investigation was always carried out by the methods developed with naturally radioactive isotopes, using the ionising action of the radiation emitted. In some cases the sensitivity of these methods was sufficient for the detection of single atoms. Thus there was a sensation when, at the beginning of the 40's, American scientists irradiated gold for months with neutrons from the concentrated source of the Berkeley cyclotron, in accordance with the reaction



and obtained the natural mercury isotope in such quantities that, after separation from the irradiated gold in a capillary tube, it was possible to observe it during a couple of minutes by the spectroscope.¹

Here however we do not desire to deal merely with the production of new chemical elements in quantities which can only be detected by nuclear physical methods, but also with methods of obtaining weighable quantities which can be used for normal chemical work. This latter objective was attained as a result of the fact that, in the uranium fission, not only are atomic fragments of moderate weight produced, but also neutrons are set free, and these in their turn offer the possibility of a "chain reaction" of enormous extent.

¹ J. H. WIENS and L. ALVAREZ, *Phys. Rev.*, 58 (1940) 11, 1005.

3. THE CHAIN REACTION

A neutron, striking the nucleus of uranium 235, causes the latter to split, during which process two or three neutrons are shot off. Thus the fission has not only, as it were, given back the original neutron, but also released fresh ones. All these neutrons can, if they are not absorbed by other substance and do not escape to the outside, cause fresh fissions in their turn when they again strike uranium nuclei. In this way, from the original fission of a single atom there may be produced in a very short time an avalanche of new fissions, a *chain reaction*, resulting in the formation of weighable quantities of the fission products.

The possibility that neutrons were set free in the fission process had been considered earlier by STRASSMANN and myself¹. Although we were not able to give an experimental proof of this at that time, the view was based on a calculation which showed that the atomic weights of the fragments must be higher than that of the highest known inactive isotopes of the elements produced. Thus the mass numbers of the highest stable barium isotope and the highest krypton isotope are respectively 136 and 86. The sum of these is 224, while the uranium which had undergone fission after taking up a neutron had the mass number of 236 (see table 1, p. 27). The primary fission product therefore had 12 neutrons too many. On the one hand, this was the cause of the radio-

¹ O. HAHN and F. STRASSMANN, *Naturwiss.*, 27 (1939) 89.

activity of the products, which passed over into elements with higher nuclear charges (and thus higher nuclear number) with emission of β -rays. On the other hand, superfluous neutrons could be emitted directly.

The actual experimental proof of the emission of neutrons during the reactions was given first by JOLIOT and co-workers in France¹, and also, simultaneously and independantly, in the United States. Theoretically this opened up the possibility of the realisation of a chain reaction and with it a practicable technical utilization of the energy set free in the fission.²

The use of this energy may be visualised in either of two ways: one comparable with the regular burning of coal in a boiler, and the other as an uncontrolled reaction developing with enormous violence, comparable with an explosion of unlimited dimensions.

In 1942 it was known in Germany and America that it was practicable, in principle, to construct power sources for the utilisation of nuclear energy. German nuclear physicists planned to develop such fission ovens.

In America, during the war, attention was concentrated on the use of the uncontrolled reaction for making explosives of previously unknown power. This is not possible with the natural isotope mixture of uranium, since, as stated above, it is the rare uranium isotope 235 which is almost entirely responsible for the fission and the neutron emission associated

¹ H. VON HALBAN, F. JOLIOT and L. KOWARSKI, *Nature*, 143 (1939) 470, 680.

² S. FLÜGGE, *Naturwiss.*, 27 (1939) 402.

with it. The uranium 238, which is present in much greater quantity, merely hinders the reaction. We shall come back to this question later. It is thus necessary to separate the small amount of uranium 235 from the large amount of uranium 238. These however, as isotopes, have the same chemical properties. Their separation can only be effected by physical methods such as diffusion, mass spectroscopy, or similar methods. These are very slow processes which require enormous plant, but in spite of that they were carried out successfully in the United States.

There is, however, another way. It consists in making use of the neutron reaction which occurs with uranium 238, that is with the commonest isotope, and leads to elements with a higher nuclear charge than uranium itself. This reaction consists in the capture by uranium 238 of neutrons having a definite, not too high, energy in a so-called resonance process. The neutrons do not however split the uranium, but produce an artificial uranium isotope with mass 239. This isotope emits β -rays, passing into element 93; from this by further β -radiation is produced a representative of element 94. The first step in the explanation of this process—the detection of the artificial uranium isotope with mass 239—was taken by MEITNER, STRASSMANN and myself:¹ the decomposition products, that is the actual elements 93 and 94, were first detected later in America. The element 93 received the name of *neptunium*, and the element 94 that of *plutonium*, from the

¹ O. HAHN, L. MEITNER and F. STRASSMANN, *Ber.*, 69 (1937) 905.

planets Neptune and Pluto which follow Uranus. (See further in chapter III, 6 and 7.)

We have suggested earlier, after the discovery of nuclear fission, that the reaction products, which we thought at the time to be transuraniums, appeared to be all fission products of uranium, that is elements of moderate weight. When actual transuraniums were discovered later in America, they did not resemble our products. The actual transuraniums were produced from normal uranium 238, while the fission products formerly considered to be transuraniums came from the rare uranium 235.

Thus in the action of neutrons on the mixed element uranium there are two concurrent processes: the splitting of the isotope 235 with emission of excess neutrons, and the capture of neutrons by the isotope 238 with formation of elements 93 and 94. Now while the neptunium isotope $^{239}_{93}\text{Np}$ has a short life time, the plutonium isotope $^{239}_{94}\text{Pu}$ produced from it has a long life. Like the uranium isotope 235, it should be possible to split it by the action of neutrons and thus obtain the material for an explosive. This conclusion is based on nuclear physical considerations, according to which an isotope with an uneven mass is more easily split than one with an even mass. The production of uranium 235 by separation from the isotope 238, which is present in much greater quantity, is, as we have seen, a very difficult problem. On the other hand, it could be anticipated that the separation of the element 94, plutonium, from element 92, uranium, would be

possible by normal chemical methods. The problem now was to produce the plutonium in larger quantities (say kilograms) in the uranium, and then to separate the new element from the uranium. This should give the material for an "atom bomb."

How would it be possible to obtain greater quantities of this material? This could be done by attaching to uranium 238 the neutrons which are produced on fission and which increase so greatly in number in the chain reaction. It is obvious that for this purpose thousands of kilograms of purest uranium metal would be necessary.

We shall come back to this in chapter III, 7.

Further research was completed in two directions. In the Kaiser Wilhelm Institute for Chemistry, the numerous reactions which occur in the fission were investigated systematically, with the result that at the end of the war we were able to make the table, referred to earlier, of direct and indirect products of the fission of uranium, arranged according to their mutual genetic relations¹.

Physical research took a different path. If it should be found possible to bring about the chain reaction, that is to cause the neutrons set free by fission to help in further splitting, then the enormous increase in fissions would bring up the energy output to measurable, and ultimately very large, amounts. The heat produced by the slowing down of the atomic fragments would cause a rise in temperature

¹ Cf. the more extended reproduction in Table 2, pp. 30—33.

in the whole mass. Two processes could now be imagined.

On the one hand a practically unlimited progressive series of fissions; the result would then be an explosion of the greatest imaginable intensity. The reactions are actually completed in an extremely short time: the speed of the neutrons set free is approximately 10^9 cm/sec. On the average these would strike a fresh uranium nucleus after penetrating merely a few centimetres of uranium, and thereby continue the chain reaction. It can be seen that already, after a millionth of a second, an enormous number of neutrons would have been formed and therefore a considerable proportion of the fissionable material would already have been split. The temperature would rise by many millions of degrees.

On the other hand, however, the process could be so arranged that the chain reaction also proceeds, but that the production of energy is uniform, regulable and continuous. This would be the case if, of the new neutrons which are produced for every fission-causing neutron, one remained available for further fission and the others were removed from the process by absorption by some other material or by diffusion. The result would be a controlled chain reaction, a kind of atomic energy machine. We shall deal later with the quantities of energy hereby produced, and with the way in which such a machine could be constructed.

Actually however the conditions are certainly not so simple as was supposed shortly after the discovery of the fission of uranium and of the neutrons set free thereby. Natural uranium

will give neither an explosion nor a source of energy, since this uranium consists of a mixture of isotopes.

4. THE DIFFERENT BEHAVIOUR OF U235 AND U238

We have already referred to the fact that natural uranium is composed essentially of two isotopes: the U238 present in great excess, and the much rarer U235, comprising about $\frac{1}{140}$ part of the isotope mixture. These two isotopes behave differently with regard to neutrons.

The rare isotope U235 is mainly responsible for the fission reactions. It is especially the slow neutrons which play a part here – that is, those neutrons which have lost part of their energy by collisions with other atoms. On the other hand, with the isotope U238, present in excess, another reaction occurs, consisting in the capture of a neutron of certain velocity with the formation of an artificial uranium isotope with mass 239 (resonance process). Already some years before the discovery of nuclear fission, LISE MEITNER and myself had discovered this capture, and together with STRASSMANN had identified this artificial uranium isotope as a β -radiating substance with a half-life of 23 minutes¹. An element with the nuclear charge of 92, which is a β -emitter must pass into a representative of the next higher element with nuclear charge 93. At the time

¹ O. HAHN, L. MEITNER and F. STRASSMANN, *Ber.*, 69 (1937) 905; L. MEITNER and O. HAHN, *Naturwiss.*, 24 (1936) 158; L. MEITNER, O. HAHN and F. STRASSMANN, *Z. Physik*, 106 (1937) 249.

we were unable to detect this substance directly with our weak sources of radiation. It was discovered in the United States, and later called neptunium¹. (see chapter III, 6). The other artificial active atoms, thought by FERMI and ourselves to be transuraniums, appeared, after the discovery of the fission of uranium, to be all representatives of the elements of intermediate atomic weight; neptunium was the first actual transuranium.

Thus, as indicated above, we are able to distinguish two different processes in the mixed element uranium: (1) the fission of U235, caused specially by slow neutrons, which results in the production of fresh neutrons and proceeds with a great development of energy; (2) the capture of resonance neutrons, of a velocity somewhat greater than those referred to under (1), by U238, without neutron surplus and without appreciable production of energy. The neutrons set free by the fission have originally, as already stated, very large energy, but this decreases very rapidly owing to collisions with the uranium itself and with other elements present. Before however reaching the particular "thermal" velocity required for the fission of U235, the neutrons pass through the stage of the "resonance" velocity which is critical for the capture by the U238 present in much greater quantity. Thus in this stage they are removed by the U238 and are not available for the fission of U235. As there is no continuous replacement of neutrons in this fission process, there can be no chain reaction.

¹ E. McMILLAN and T. H. ABELSON, *Phys. Rev.*, 57 (1940) 1185.

5. THE URANIUM PILE

In the solution of the problem of avoiding the removal of the extra neutrons by the U238, the work of nuclear physicists was in the foreground. One possibility would appear to lie in the use of certain substances which reduce the speed of neutrons below the critical capture speed without absorbing them to any appreciable extent. These substances are called *retarders* or *moderators*. If now a construction is made with the moderator and the uranium in a sort of space lattice arranged in such a way that a uranium layer is always surrounded by a layer of moderator, then the fast neutrons formed in the uranium layer come in due course into the surrounding moderator. Here their speed is reduced below the dangerous resonance velocity, so that they pass with this lower velocity into the next uranium layer, where they are able to split a fresh lot of U235.

Especially suitable as moderators are *heavy water* and purest *graphite*. In Germany heavy water was chosen (separated on a technical scale from natural water in occupied Norway), that is water which contains, in place of the ordinary hydrogen isotope of mass 1, the heavy isotope with mass 2, and which, unlike ordinary water, hardly absorbs neutrons but merely reduces their speed. In the U.S.A. pure graphite was used instead, as it was more easily obtainable in very large quantities.

In 1942, investigations had advanced to about the same point in Germany and in America. It was known that ~~it was~~

possible, in principle, to make not only a source of heat and energy for peaceful purposes but also a bomb with a previously unknown destructive power.

One possibility was the separation of U235 from very large quantities of ordinary uranium; in U235 all the neutrons always have a splitting, that is an energy-giving, action. This separation of the rare U235 from the 140-fold excess of U238 could be done only by physical methods and would have been impossible in Germany during the war owing to the lack of technical means.

The other possibility for the production of an "atom" bomb was based on the capture of neutrons of a definite speed by U238, as described above. From the artificial U239 formed in this way there is produced, as already mentioned, a transuranium with nuclear charge of 93, neptunium, which emits β -rays and thereby forms the element 94, plutonium. This element of atomic weight of 239 is a fairly stable substance; it is formed in the energy-producing atomic plant mentioned above, called in the U.S.A. the *uranium-pile*, in England the *atomic pile*, and in Germany *Uranbrenner*.

There was however a long way to go from the point at which the possibility of making practical use of the energy was seen in principle, to that at which the construction of an energy-giving installation or – as a further consequence – an atom bomb became a fact. In the first place it was necessary to bring about a steady controlled chain reaction; a process sharply defined by three possibilities. For every neutron that

has split a uranium atom, only one of the extra neutrons released thereby must produce a further splitting, in order to attain a stable process. It is of vital importance that this should be exactly one neutron; if a little less, then the chain reaction is broken and the machine stops, or in other words, the oven goes out; if a little more, then the reaction is no longer under control, and the rapid increase in the number of fissions leads to explosion. Possibly the force of such an explosion is not to be compared with that of an atomic bomb, still the result would be to destroy the installation.

The conditions essential for a smoothly functioning and non-exploding machine may be actually created on the basis of two phenomena. In the first place *cadmium* metal has the property of strongly absorbing neutrons. By the use of moveable cadmium bars it is possible to remove excess neutrons from the system. Further, it appeared—what is very important in this connection—that there is a certain *delay* in the emission of a small proportion of the neutrons set free by the fission. This makes it possible that any regulation accomplished with the aid of cadmium would not have to be carried out in the fraction of a second, but could be done after a few minutes. In place of cadmium *boron* may be used¹.

6. PLUTONIUM

While after 1942 the work of physicists in Germany was limited to the development of an energy-producing machine,

¹ For further details see J. CHADWICK, *Nature*, 159 (1947) 421.

that is an atomic pile, in America, besides the work on the pile, all available resources were used to make the atom bomb, more than 2,000 million dollars being invested in the project¹. This objective was attained partly by the preparation of the isotope U235, partly by the chemical separation of plutonium, which is produced continuously from the irradiated uranium in the pile. It was in fact to be anticipated that this plutonium could be split by neutrons of any arbitrary energy. For the purpose many tens of thousands of kilograms of purest uranium were necessary, and even larger quantities of pure graphite as moderator; it was also necessary to have a plant for the separation of the plutonium formed from the uranium, and this had to be controlled from a safe place not exposed to the danger of radiation. The rays from the artificial fission products which are produced continuously in the pile are so enormously active that it is impossible to approach the neighbourhood of the pile or of the separating plant without danger.

It was indeed a wonderful performance when American and English chemists and physicists together succeeded, within a few years, in determining accurately the chemical properties of the artificially prepared new element 94, plutonium, using only fractions of a milligram. This work was so accurate that it was possible to build enormous installations for the manufacture of kilograms of this substance on the basis of these observations, and that the final objective was attained¹. With justified pride, these American workers announced a

¹ See further Chapter III, 7.

production increase with a factor of $1 : 10^{10}$, i.e. 1 to a 1,000 millions!

After hearing of the danger involved in working-up the plutonium formed in the pile, owing to the powerful radiation from the installation, one would imagine that working with plutonium itself, that is with the material of the bomb, would be highly dangerous and require the greatest caution. Actually this is not the case. Pure plutonium in not too great quantities is quite harmless. The β -rays which it emits are practically completely absorbed by a thick piece of paper or by aluminium foil.

The dangerous rays in the pile come from the numerous fission products, the formation of which – with emission of extra neutrons – makes the chain reaction possible. Once these products are removed from the plutonium by chemical separation processes, then working with plutonium has, up to a certain limit, no particular danger.

7. CONTROLLED AND UNCONTROLLED REACTION; THE ATOM BOMB

There is, however, a certain critical quantity above which plutonium is dangerous. If this quantity is exceeded, that is if there are more neutrons in the material than pass away to the outside, then we have the terrible explosive whose action we have learnt to know in the form of the atom bomb. The

same holds also for pure U235. Thus explosion occurs when the mass of plutonium is so great that the few free neutrons always present are able to initiate a chain reaction. We do not know exactly how great this critical quantity is: possibly it is 10, perhaps 20 or 30 kilograms, but certainly not much more. The explosion can be released by, for example, bringing together at great speed two half-spheres of plutonium, each being in itself a harmless quantity. The conditions for the chain reaction are then present, and explosion follows within a fraction of a second.

By the summer of 1945, three bombs had been made in America. One was exploded in a "practical test" in New Mexico; one U235 bomb fell on Hiroshima; one plutonium bomb on Nagasaki. The result is well known: the war with Japan was ended. Since then the Americans have undoubtedly made hundreds of kilograms of plutonium in their gigantic uranium piles.

In Figs. 1 and 2 (pp. 61 and 62) the chain reactions are shown diagrammatically. Fig. 1 shows the controlled reaction in the atomic pile. It is assumed that in the fission three neutrons are set free: one is lost by diffusion to the outside or by absorption; a second is captured by the U238 and forms plutonium; and the third avoids these processes and splits fresh U235, thereby supplying the "ammunition" for the continuation of the chain reaction.

Fig. 2 shows in comparison the uncontrolled reaction. U238 is absent; plutonium or U235 separated from U238

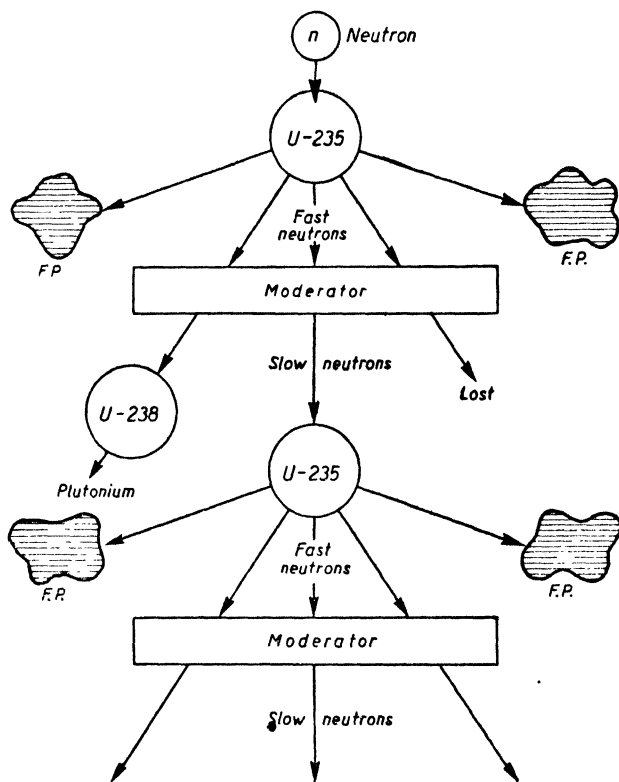


Fig. 1. Controlled reaction in the uranium pile

Of the three neutrons set free in the disintegration of $U235$, one is lost by diffusion or absorption, a second is captured by $U238$ and forms plutonium, the third splits a new atom of $U235$ and thereby keeps the chain reaction going.

F.P. = fission product

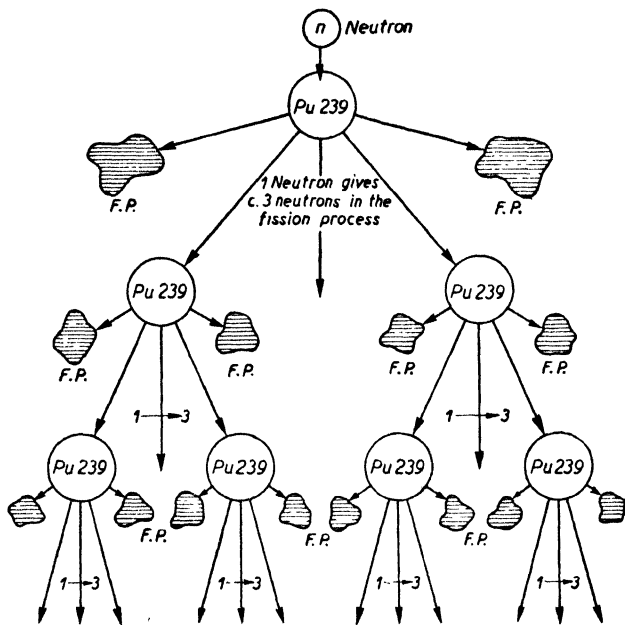


Fig. 2. Explosive, uncontrolled reaction in the atom bomb

U238 is absent. The disintegration occurs in plutonium or U235. The speed of reaction is enormous, consequently the bomb explodes.

F.P. = fission product

acts as fissionable substance. The reaction shown here proceeds with the greatest imaginable velocity, and the explosion of the bomb is the result.

In place of the captions "controlled" and "uncontrolled" chain reaction under Figs. 1 and 2 respectively, one should

perhaps write the words "hope" and "fear". Can we *hope* that nuclear energy will be adopted for the benefit of progress and of mankind, or must we *fear* that it will be realised in more numerous and larger atom bombs, producing fear and anxiety?

Perhaps nobody can answer these questions to-day. One can however easily imagine what a future atom bomb war would mean for harassed mankind . . .

8. THE HYDROGEN BOMB

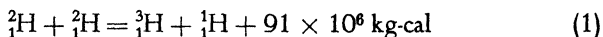
Remembering the effect of the atom bombs on Hiroshima and Nagasaki in August 1945, or considering the investigations at Bikini in 1946, one would think that mankind had already carried it magnificently far enough with the utilisation of atomic energy for destructive purposes and that there would be no desire to want to add still more powerful ones to these means. Nevertheless this seems to be the case. Since the end of 1949 there has been talk of the "hydrogen bomb", and President TRUMAN has ordered the development and construction of the "hydrogen bomb" to be officially begun in order to create a new weapon for keeping the world peace.

What is to be imagined by a "hydrogen bomb"?

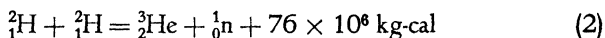
Through the calculations of VON WEIZSÄCKER in Germany and BETHE in the U.S.A. a clear picture can be formed of the nuclear processes which go on in the interior of the sun. With the participation of carbon as "catalyst", hydrogen is

slowly built up into helium. For this, it is assumed that in round figures a temperature of 10 to 20 million degrees is necessary; such temperatures appear to occur in the interior of the sun. By this slow synthesis of helium with mass 4 from ordinary hydrogen of mass 1 it has been possible for the sun to maintain its temperature for several thousand million years and to enable life to exist on the earth for about the same length of time.

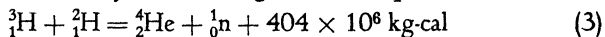
Thus the building up of helium from light hydrogen is, in principle, quite another process than the break-down of the heavy elements uranium or plutonium into lighter elements which takes place in the atomic bomb. In the centre of the atomic bomb at the moment of explosion, temperatures are thought to occur which certainly reach millions of degrees and which consequently are not too far from the temperature of the helium synthesis. Now, in addition to ordinary light hydrogen of mass 1, "heavy" hydrogen of mass 2, *deuterium*, is also known. From two atoms of deuterium, hydrogen of mass 3 (*tritium*) and helium of mass 3 can be synthesized according to the equation:



and



Normal helium can be made from tritium, which can be prepared synthetically, according to the equation:



The energies set free per unit weight in the transmutations

of ${}^2_1\text{H}$ and ${}^3_1\text{H}$ are comparable with the energies liberated in the fission of U235 or Pu. However, for the reactions (1) to (3) to come into action the ${}^2\text{H}$ - and ${}^3\text{H}$ -nuclei must bombard each other with such speeds that the repelling electrical forces are overcome. At a temperature of some million degrees the thermal speed of the hydrogen atoms is so great that the nuclei come into contact with each other and hence the nuclear reaction can take place. The enormous energy liberated in this way raises the temperature, thus accelerating the transformation, i.e. an explosion-like conversion of the whole deuterium and tritium energy.

Hence in order to initiate this explosion, an ignition temperature at only one spot of about 20 million degrees must be attained. This temperature, however, is probably available at the centre of an exploding uranium bomb. Thus according to the theoretical calculations, one can construct a bomb consisting of ${}^2\text{H}$ and ${}^3\text{H}$ and containing a uranium bomb as detonator.

The great "advance" of this H-bomb is that there is no upper limit for it. Its effect can be made as great as desired. It is only dependent on the amounts of deuterium and tritium which are available.

The particularly unpleasing thing about the hydrogen bomb is that it will never be possible, as in the case of the fission of uranium, to utilise the nuclear energy of the hydrogen reactions for peaceful purposes. We can reach the temperature of 20 million degrees or more only for millionths of a second

and not for any length of time. A "controlled reaction" is not possible.

The same nuclear reactions which have been going on in the sun for millions of years and which yield the energy forming the basis of our life on this earth, become in the hands of man simply a means of destruction and nothing more.

9. ADAPTATION OF THE CONTROLLED REACTION

We will now limit ourselves to the symbol of hope, and consider briefly the possibilities which the utilisation of nuclear energy will offer for civilization. There is no doubt that the atomic piles in use in America are merely the forerunners of technically much more perfect "uranium machines". The uranium pile of to-day already however indicates quite clearly the two most important fields of employment of the controlled chain reaction. These are, on the one hand, the use of the energy in the form of heat or electricity; and on the other hand the production of artificial radioactive chemical elements with their numerous possibilities of application in various spheres of pure science, biology, medicine and technology. Moreover, the atomic pile is an extremely powerful source of neutrons, so that the study of the behaviour of these neutrons, both fast and slow, forms in itself a wide sphere for extremely important fundamental research.

Let us consider first the production of energy. A simple calculation shows that the energy set free in fission reactions

is millions of times greater than that of the most powerful chemical reactions.

The energy which an electron possesses after acceleration by a potential difference of 1 volt is called an electron volt, abbreviated eV. As this quantity is very small, it is usual in nuclear physics to work with a quantity one million times this, the MeV. 1 MeV corresponds to the energy of 1.6×10^{-6} ergs. Expressed in heat units, $1 \text{ erg} = 2.39 \times 10^{-11}$ kilogram-calories. In the fission of uranium the fission products receive a kinetic energy, per uranium atom, of at least 160 MeV.

Thus, starting with 1 kg of plutonium (or 1 kg of pure U235), it is easy to calculate the quantity of energy set free by its controlled or uncontrolled decomposition:-

239 g Pu (239 = atomic weight of Pu) contains 6×10^{23} atoms; 1 kg contains 2.5×10^{24} atoms. Per atom of Pu, 160 MeV are set free, thus per kg $2.5 \times 10^{24} \times 160 = 4 \times 10^{26}$ MeV. As 1 MeV corresponds to 1.6×10^{-6} ergs, therefore 4×10^{26} MeV are equivalent to 6.4×10^{20} ergs. As moreover 1 erg corresponds to 2.39×10^{-11} kg-cal, the heat set free on the decomposition of 1 kg of plutonium is equal to $2.39 \times 10^{-11} \times 6.4 \times 10^{20} = 1.5 \times 10^{10}$ kg-cal. The heating power of 1 kg of good coal is barely 8000 kg-cal. Thus the production of energy on the complete disintegration of 1 kg of plutonium appears to be 2 million times as great as that obtained from 1 kg of coal! In practice this amount must be increased still further by the energy of transformation of the numerous fission products.

These figures show the enormous heat and pressure waves which must occur on the explosion of about 10 kg of plutonium, even if only 20% of the material is actually split and the other 80% is lost during the explosion by evaporation.

In the regular chain reaction in the atomic pile, the fission process is stationary. When using the natural mixed element (U238, U235) only the U235 is split, as mentioned above. The quantity of energy set free thereby is practically the same as with plutonium.

If, in a large atomic pile, 1 kg of U235 is split per day, then the daily production of heat is the above-mentioned 15,000 million kg-cal. Possibly the fission ovens used in the U.S.A. for the manufacture of the first large quantities of plutonium were not so large that 1 kg of U235 was used up every day, but they could not have been much smaller.

Even with smaller piles it is a difficult problem to remove the heat of reaction, so that the temperature of the installation does not rise too high. From published results it may be taken that the installations in use at Hanford (in the North-West of the U.S.A.) were adjusted to a temperature of not more than 100° C. The excess of heat was carried away by the water of the Columbia River, which has approximately the same width and depth as the Rhine, and this water rose in temperature by about 1°. The heat which was not utilised would have been sufficient to provide a town of about 50,000 inhabitants with central heating.

From these results it is obvious that it is merely a question

of a short time before the heat set free in the atomic pile is made available for practical purposes, for example, for the supply of hot water to places and factories from a central station.

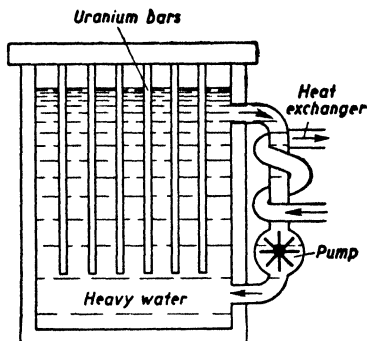


Fig. 3. Diagram of an atomic pile (fission oven) as source of heat.
Moderator: heavy water

Fig. 3 shows, in a much simplified manner, how an atomic pile using heavy water as moderator can be imagined as a source of heat. The heavy water which surrounds the holders of the uranium bars becomes strongly heated, and gives up its heat continuously in a heat exchanger to the medium used for the central heating.

A more difficult problem, not yet solved, is that of converting the heat into electricity with a favourable efficiency. A heat source of no higher than 100°C can only be transformed very inadequately into electrical energy: much higher temperatures

are required. At the moment, however, temperatures above 300° C for a uranium pile in action are not to be anticipated: not so much because the pile would not be able to produce enough heat, but because the aluminium, used as mutual screening of the uranium and graphite, will not stand much higher temperatures: also because the corrosion of the uranium by the higher temperature and the intensive radiation would become too great, as well as for a number of other reasons. It is not possible to foresee when these technical difficulties will be solved. That they will be solved, one can hardly doubt.

It is easy to calculate the electrical energy corresponding to the above-mentioned amount of heat and an efficiency of 100%. 1 kg-cal is equivalent to $\frac{1}{860}$ kWh; the heat produced by the splitting of 1 kg of U235 or of Pu is 15,000 million kg-cal, which corresponds to

$$\frac{1.5 \cdot 10^{10}}{860} = 1.75 \cdot 10^7 \text{ kWh}$$

or approximately $\frac{3}{4}$ million kW days. For thermodynamic reasons, quantitative conversion of the heat into electric current is impossible, even with a high operating temperature. But even with an efficiency of about 60% the amount of energy available is about equal to that of a large power station.

With large piles there are further difficulties in addition to that of the conversion of the fission energy into electrical current. The chief one is the danger from the intensive radioactive radiation with which the action of the pile is inevitably

associated. By the fission of 1 kg of U235, about 999 g of active elements are formed, while about 1 g is converted into radiation energy. This 999 g of active elements corresponds, with respect to β - and γ -radiation, to about one million kilograms of radium.

The same amount of radiation is produced by the exploding bomb, but here however the largest part of the active elements rapidly decomposes. Many fission products have a life of merely a few seconds, minutes or hours. These substances of short life are produced continuously in the pile and, assuming the disintegration of 1 kg of fission products per day as above, the resulting activity corresponds to many tons of radium. It is for this reason that very thick protective screening walls are necessary to protect the neighbourhood from this radiation, and also from the many neutrons which diffuse out of the pile.

The difficulty in constructing this protective screen round the atomic pile is due to the simultaneous emission of γ -rays and neutrons. Heavy metals—iron or, better, lead—are necessary for the absorption of γ -rays, but the neutrons are little affected by this. They need for their absorption light hydrogen-containing substances, which however only absorb the γ -rays very slightly.

Thus a pile with the relatively small dimensions of 1 cubic metre would require more than 100 tons of screening material. The total weight of an installation of the future, has been estimated by CHADWICK as some thousands of tons.

Fig. 4 shows a diagram of a hypothetical experimental installation for obtaining electric current with the aid of a steam turbine driven by the heat of an atomic pile¹.

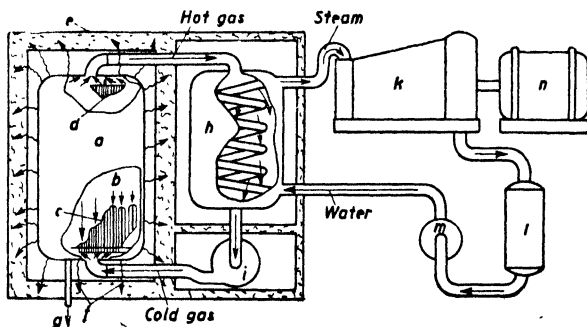


Fig. 4. Diagram of an (imaginary) installation for producing electric current with the aid of a steam turbine driven by the heat given off by an atomic pile.

- | | |
|------------------------------------|-------------------------------|
| <i>a</i> Atomic pile | <i>h</i> Cooler |
| <i>b</i> Holders with uranium bars | <i>i</i> Gas circulator |
| <i>c</i> Graphite as moderator | <i>k</i> Turbine |
| <i>d</i> Cadmium control bars | <i>l</i> Condenser |
| <i>e</i> Protective covering | <i>m</i> Pump |
| <i>f</i> γ -rays | <i>n</i> Electrical generator |
| <i>g</i> Fission products | |

In place of the heavy water of Fig. 3, which served as moderator and for heat transport, we have here a gas, probably helium, for the heat transfer. On the left of the figure may be

¹ Taken from CHAPMAN PINCHER, *Atomic Age Exhibition*, London, 1947.

seen part of the interior of the furnace, called here atomic pile. Around the holders *b* for the bars of uranium circulates the gas, which is introduced cold below. The gas channels are surrounded by the moderating graphite *c*. At *d* may be seen one of the control rods, composed of cadmium, which strongly absorbs neutrons (cf. section 5 above.) Below is an outlet *g* for the fission products. As these latter are produced in the uranium and accumulate there, this opening *g* forms a place from which the irradiated uranium bars may be removed and replaced by fresh ones. Around the whole pile is the screening *e* to absorb γ -rays and the neutrons which diffuse out.

In Table 4 the separate processes, which occur simultaneously in the atomic pile, are collected together¹.

Table 4. SUMMARY OF THE PROCESSES OCCURRING IN THE URANIUM PILE

Process	Chain reaction	Process in pile	External effect
A Nuclear fission	Is the source of fresh neutrons and therefore keeps the chain reaction going	Leads to the formation of radioactive substances and thereby also to strong radiation	Has as consequence that the pile functions as source of energy

¹ Taken, in somewhat modified form, from H. THIRRING, *Die Geschichte der Atombombe*, Vienna, 1946, p. 104.

Process	Chain reaction	Process in pile	External effect
B Emission of neutrons to exterior	Decreases the number of neutrons and therefore acts as a brake on the chain reaction	—	Results in the pile acting as strong neutron source
C Capture of neutrons in the uranium nucleus without fission	As B	Formation of $^{239}_{92}\text{U}$, from which is produced by β radiation $^{239}_{93}\text{Np}$ and $^{239}_{94}\text{Pu}$ (the explosive of the atom bomb)	—
D Capture of neutrons in nuclei other than uranium	As B; can be used for the control of the chain reaction by means of moveable Cd control bars	—	—

10. ARTIFICIAL RADIOACTIVE ELEMENTS

a. Production

We have already given a summary (Table 2, pp. 30–33) of the numerous artificial radioactive elements which can be detected after the fission of the uranium atom. This fission always proceeds in such a way that two elements of moderate weight are produced, their nuclear charges amounting together to 92 (= the nuclear charge of uranium).

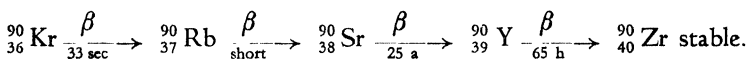
The active isotopes, sometimes numerous, belonging to a particular element are characterised among themselves by their half-life times which are often very different, that is, by their differing stabilities. We have already met substances with half-lives of merely a few seconds or of fractions of a second, also some with half-life times of many years.

All the active fission products, in so far as they have a sufficiently long life, and all the inactive end products of the chain reactions—about 70—can now be produced, or will be in the near future, in the atomic pile in weighable amounts; as also can representatives of all elements which can be formed by irradiation with neutrons. In the atomic pile we have acquired an extremely important source of neutrons for the production of such substances.

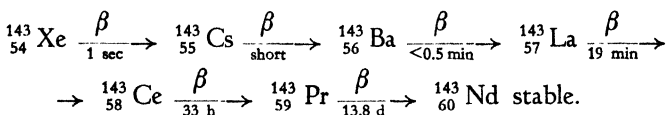
We cannot give here a description of the many kinds of active atoms which are produced directly and indirectly in the uranium fission. The large number of isotopes of xenon and

krypton is very striking. They are mostly substances of very short life which, like the other fragments, generally continue to change by way of β -emission intermediate products— isotopes of higher elements—until they reach an inactive state corresponding to a naturally occurring stable atom of the same atomic number. The direct fission products, that is the first terms of this series, have in fact a mass too great for their nuclear charge, therefore they move up to higher nuclear charges.

We will illustrate this by two examples taken from the already mentioned inert gases xenon and krypton.



The atom which appears as krypton from the uranium fission has a mass of 90 and nuclear charge of 36, and only becomes stable with charge of 40, i.e. as zirconium. A similar case occurs with the atom of mass 143 which appears as xenon with charge 54. Only after passing through five other intermediate products does it become stable, as neodymium, with a nuclear charge of 60.



As may be seen from Table 2, similar transformation series also appear in many other cases.

Other important active atoms which appear in the uranium

fission are isotopes of the element 43, which does not occur naturally and is a homologue of manganese (see Chapter III, 2); and isotopes of the yet unknown rare earth, element 61 (see Chapter III, 3). These representatives of new artificial chemical elements of long life can be obtained in weighable amounts in the atomic pile. This is especially the case for the element 43, whose isotope with mass 99 has a half-life of 4 million years. Thus in practice this substance can be considered as a stable chemical element.

In the atomic pile are produced, not only those kinds of atoms which are formed directly as the result of the uranium fission, but also all artificial active elements and kinds of atoms which result from the action of neutrons on ordinary elements. And there are a large number of these¹. In the pile neutrons are produced in the highest imaginable density. Many of them diffuse outside into the surroundings of the pile and may be utilised there (see Table 4). The elements to be irradiated may also be placed in the pile by means of probes, so that the irradiation occurs in the region of highest neutron intensity. In order to compensate for the loss of neutrons the absorbing cadmium rods must be partially withdrawn from the pile.

In this way, in addition to the fragments of intermediate mass, the lighter and heavier elements also may be made artificially active. Of special interest are active carbon (of long life), sodium, phosphorus, calcium, etc.

¹ See for example G. T. SEABORG and I. PERLMAN, *Rev. modern Physics*, 20 (1948) 585.

b. Application

In practically all spheres of chemistry, whether pure science, applied chemistry, physical chemistry, or biology, the artificial active elements can be of use on account of their ease of detection, even in unweighable quantities. Here we will limit ourselves to some questions of metabolism. The fundamental work on this subject is due mainly to HEVESY¹.

Radioactive phosphorus has already been used in many ways in investigations of problems of the metabolism of plants and animals; phosphorus is an essential constituent of living material. The speed of assimilation and the exchange of fresh phosphorus atoms for ones already deposited in the organism, including the exchange in hard structures such as teeth and bones, can be studied in detail with the aid of the "marked" or "labelled" phosphorus. It has also been used successfully against leucaemia. With different preparations and application on a broader basis, it is hoped that a permanent success will be obtained.

Radiostrontium has been used for certain forms of joint carcinoma and other diseases.

Radio-iodine has been known for a long time as a treatment for certain thyroid complaints. Iodine is a constituent of the thyroid gland, and certain diseases of the latter are due to a shortage of iodine. The speed of absorption of the iodine in

¹ See for example G. SCHUBERT, *Kernphysik und Medizin*, Göttingen, 1947; G. HEVESY, *Radioactive Indicators, Their Application in Biochemistry, Animal Physiology, and Pathology*. New York and London, 1948.

connection with the iodine preparation used, and the quantity taken up in comparison with that in the blood circulation, can be accurately determined. For this purpose the GEIGER-MÜLLER counter¹, so valuable for the detection of individual radiating particles, is held outside the body under treatment and shows the route followed by the active substance. The number of counts also gives an idea of the quantitative conditions.

If it is desired to use active indicators in order to find in what form chemical compounds are deposited in particular members of the body, e.g. in cases of carcinoma, then the corresponding chemical compound may be prepared in radioactive form. This may be made so strong that the carcinoma may be destroyed without appreciable damage to healthy cells (thyroid carcinoma).

As a demonstration, one may for example make visible the absorption of salt into the blood, by replacing part of the sodium in NaCl by *radiosodium*. If a draught of water, containing this radioactive salt, is taken, then the speed of transference of the salt through the intestinal wall into the blood and the different parts of the body may be followed by means of the GEIGER-MÜLLER counter. A few minutes after drinking the solution, the sodium chloride can already be detected in the fingers.

An important role has also been played by *active carbon* of mass 14 and with long life. Various organic chemical reactions

¹ Cf. A. TROST, *Zeitschrift des V.D.I.*, 85 (1941) 829.

may be studied with the aid of this labelled carbon. Possibly it will also bring nearer a solution of the greatest problem, the formation of chlorophyll in the green plant, and in this way enable us to discover the way in which the plant is able to synthesize organic compounds from CO_2 and water.

A large number of artificially radioactive isotopes have been prepared for scientific and medical purposes, among other places in the U.S.A., England, France, Sweden and Holland. Owing to the extremely small activity which is needed in most of the investigations, using modern methods of counting, the quantity of activity obtainable in the form of artificial elements from a single atomic pile of moderate size is practically unlimited.

11. POSSIBILITIES OF THE FUTURE

How difficult it was, after the value of powerful radium preparations for medical and scientific purposes had been proved, to obtain radium in sufficient quantities to satisfy the most urgent needs of hospitals and laboratories! Perhaps in the future the problem will be the opposite: how, when a large number of plant for obtaining heat and electricity on an industrial scale are in use, will it be possible to guard against the danger of the strong radiation from the fission products? For the latter must be removed from the uranium after it has "worked" for a certain time. Although most of the fission products break down rapidly, yet there are always more than

enough of the long-living ones remaining over. Separation of the fission products which gradually accumulate, the "ash" as it were, is necessary. As a result of their strong absorption for neutrons these act as a poison towards the continuance of the chain reaction, and above a certain concentration they would bring the machine to a standstill.

In itself, an installation of about 100 tons of uranium with a capacity for heat production corresponding to 100,000 kW, should be able to work for about 20 years without renewing the "fuel" ¹. Actually, however, the process comes to an end earlier on account of the effect of the fission products and the attack of the strong radiation on all the materials used. Thus after a certain time cleaning and renewal of the installation is necessary, and the uranium bars have to be automatically replaced by new ones. This is necessary also because the mixed element uranium gradually becomes poorer in the U235 which keeps the chain reaction going.

Meanwhile, however, the—also fissionable—plutonium is produced by the capture of neutrons by the main isotope U238. The plutonium may be added to the U238 in place of U235, after the uranium has been freed from the neutron-absorbing "ash". Thus the machine, as it were, renews its fuel from its own stock, and this proceeds until finally all the U238 has been used up.

Fig. 5 gives an instructive picture of a large atomic energy works of the future, in which is shown, in addition to the

¹ J. D. COCKCROFT, *Nature*, 160 (1947) 451.

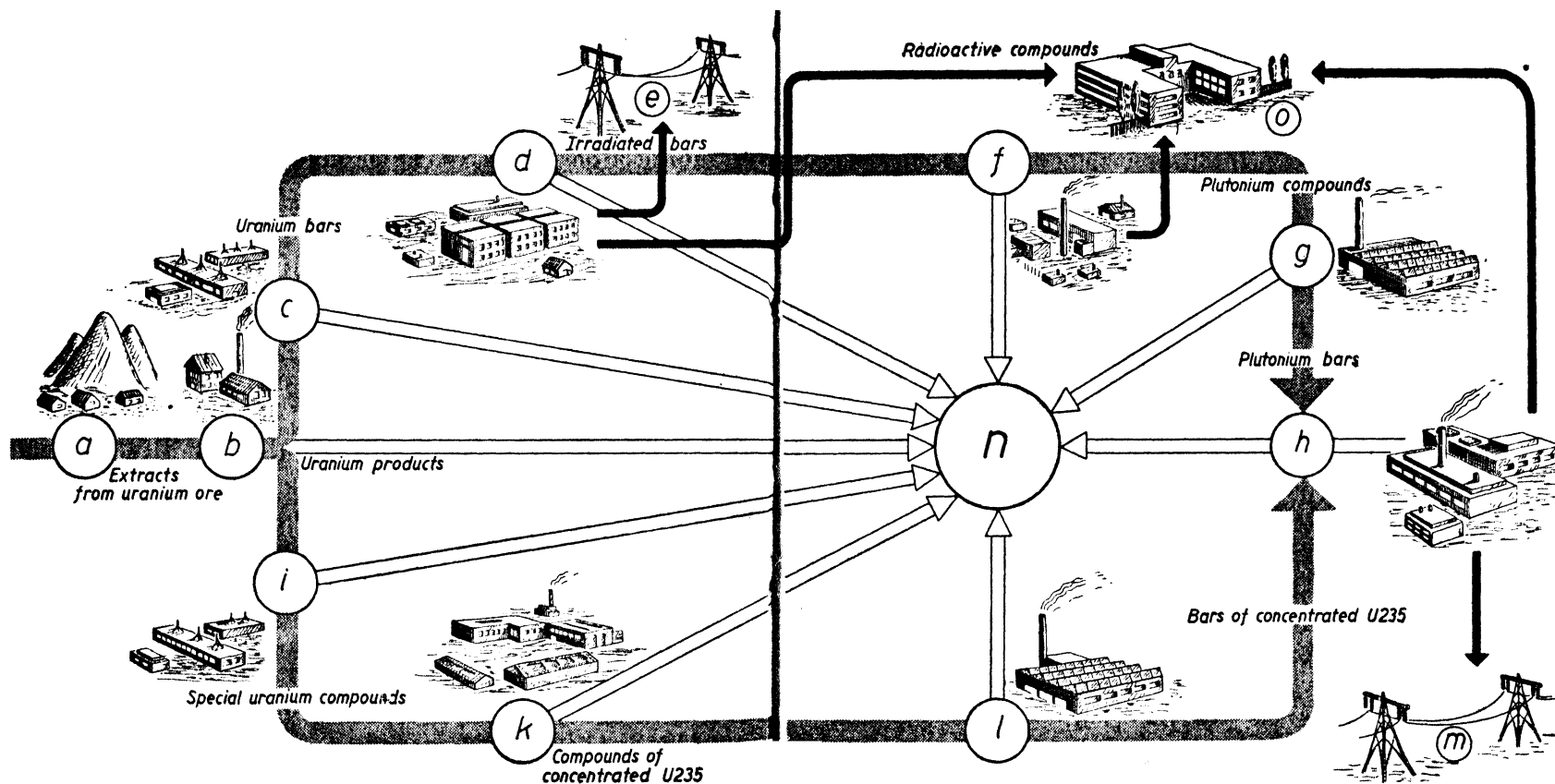


Fig. 5. Illustration of a large atomic energy plant of the future. In addition to the use of the ordinary mixed element uranium in the uranium pile, there is shown a second pile working with concentrated U235 or with plutonium.

- a Uranium mine
- b Refinery for crude uranium
- c Chemical and metallurgical works
- d Complex of uranium piles for preparation of plutonium
- e Power station

- f Factory for the separation of the plutonium formed and the fission products
- g Preparation of pure plutonium
- h Second pile complex
- i Chemical works
- k Separating plant for isotopes
- l Manufacture of bars from pure or concentrated U235
- m Second power station
- n Possible subsidiary for making secret weapons
- o Factory for making radioactive compounds

use of the ordinary mixed element uranium in the piles of installation *d*, also that of the concentrated U235 or of plutonium in the secondary installation *n*¹.

The process commences in the uranium mine *a*. In the figure, following first the upper branch, we come to the preparation of the pure mixed element in the factory *c*, from whence in *d* is made plutonium. Thus *d* is the primary main fission oven. The heat of reaction of the fission is used to provide the power plant *e* with energy. At *f* is the factory where the plutonium formed and the fission products are separated. Proceeding further, we come to *g*, the purification of the plutonium which is used on the one hand for the manufacture of atom bombs (at *n*), and on the other hand in the installation *h* (secondary fission oven).

Following the lower branch from the left, we come first to the factory *i* where the uranium is converted into the volatile chemical compound uranium hexafluoride, which is necessary for the separation of the isotopes U235 and U238; then comes the very large isotope separation plant *k*, and the preparation of bars of pure or concentrated U235 in *l*. This isotope, as also plutonium, can then be mixed with a smaller quantity of ordinary uranium and used as a further source of energy in *h* (the secondary uranium pile). The separated U235 and the plutonium freshly formed in *h* can however also be used for the manufacture of the bomb.

¹ United Nations, Dept. of Public Information, *The Control of Atomic Energy, Lake Success*, 1946.

Regarding the cost of future installations for obtaining electric power, no reliable figures can be given. It is not yet known what sort of construction will be required when working at the temperature necessary for profitable conversion of the heat into electrical energy. If this technical difficulty could be overcome, then according to American calculations the cost would be only about 25% higher than that of a power installation using coal as fuel. Possibly the cost would be reduced after further technical improvements, so that atomic power could become a competitor of coal and mineral oil ¹.

Undoubtedly such installations will be employed successfully in places where coal or oil is not available or only obtainable with difficulty (polar regions, deserts, etc.). Naturally the possibility has also been considered of using atomic power to drive large ocean liners and warships which must be able to keep going for long periods without taking in fresh fuel. Smaller ships, aeroplanes and trucks do not come into the question on account of the danger of radiation and the heavy screening necessary.

If one asks how much uranium will be available in the future for producing energy, then it must be stated that, with the quantities available in the richer uranium deposits known to-day, it would certainly not be possible to replace the present-day sources of energy (coal, mineral oil, water power). There are however large quantities of minerals and ores poor in uranium, and since at the moment uranium is approximately

¹ J. CHADWICK, *Nature*, 159 (1947) 421.

the most costly chemical element, undoubtedly methods will be developed for making use of deposits of low uranium content.

In a paper by YOUNG¹ there is an estimate of the energy which could be obtained from our deposits of coal, and from those of uranium known at present, compared with the heat which reaches the earth from the sun. According to this calculation the amount of heat which the earth receives per second from the sun is 1.7×10^{14} kW. With the same rate of production of energy the coal reserves would be sufficient for 15 days, while known rich uranium deposits would only last for 3 minutes. If on the other hand allowance is made for the total uranium content of that part of the earth's crust which is accessible to us (uranium is present in very small concentration in every mineral) then the value is 30,000 years.

If, to what has been said regarding the possibilities of a peaceful use of atomic energy, we add a brief summary, then we can state as follows:-

(a) It is possible that the heat set free in the controlled chain reaction will be available within a few years for the provision of hot water for large factories or even for towns.

(b) The provision of power stations for the supply of electric power lies further in the future. This problem will however be solved when the existing difficulties, which are merely ones of apparatus and not of principle, are overcome. Only then can one give with certainty a decision regarding the economy of nuclear energy as compared with sources of energy in use at present.

¹ G. YOUNG in: *One World or None*, London, 1947, p. 52.

III. ARTIFICIAL NEW ELEMENTS

1. INTRODUCTION

The chemical elements we are about to consider here fall into two groups—a first group consisting of the artificial new elements 43, 61, 85 and 87, which have recently filled the gaps in the Periodic Table of elements, and a second group comprising the artificial new elements 93, 94, 95, 96, 97 and 98, from that region of the Periodic Table which lies beyond uranium, hence called the transuraniums. These ten elements differ from previously known elements in that they are all artificial, having been made by man in his recent epoch-making advances in the realm of nuclear physics.

Supplies of these new elements are gradually becoming available to the ordinary research worker, and the fact that the situation in this respect is rapidly improving is an indication of a new trend—almost a “volte face”—in modern chemistry. A few years ago, for example, I gave a lecture on the subject: “*From the weighable to the unweighable in chemistry*”. In that lecture¹ I described the ever-growing refinement of means and methods at the disposal of the chemist: sensitive balances, spectrum analysis, micro-chemistry etc. enabled him to detect smaller and ever smaller amounts of elements and

¹ O. HAHN, *Mitt. Universitätsbund Marburg*, (1936) 41.

compounds; radio-active methods took him a tremendous stride forward into the region of the unweighable; until finally the WILSON cloud chamber, the GEIGER-MÜLLER counter and other instruments enabled him to detect the presence of an individual atom. Further than that he could not go. He had reached the absolute limit.

To-day we witness a movement in the opposite direction, which can be indicated by: "*From the unweighable to the weighable in chemistry*". We shall see how it is now possible to produce artificial elements in quantities amenable to chemical analysis, how grammes, even kilogrammes of substances were obtained which until a few years ago were available as invisible traces only. This enormous increase in output is directly due to the discovery of uranium fission and to the subsequent construction, first in the U.S.A. and then in other countries, of "uranium piles".

In order that such increased output may be possible it is necessary that the production process be based upon a *chain reaction* of the kind taking place in a uranium pile, since it is only by such a process that the artificial elements are produced in sufficiently large amounts. Another condition is that the products must have relatively long lives, thus permitting of sufficient accumulation in spite of competing radioactive decay.

Before proceeding to deal with individual representations, however, it should be understood that completely stable representatives of the elements to be described cannot possibly

exist. This fact was first formulated by MATTAUCH in the isobar-rules named after him (later called the "MATTAUCH isobar-principles") and afterwards shown by him to be theoretically well-founded. For our purposes the law can be stated in the following form: There can be no stable isobar-pairs (isobars = different chemical elements with the same mass-number) whose nuclear charges differ by a single unit. As an example let us take the first element on our list—element 43. All the mass-numbers that could be assigned to element 43 are already represented by stable isotopes of the neighbouring elements above and below. There is, therefore, no room left for a really stable isotope of element 43.

In fact all the artificially produced new elements are radioactive. Some of them undergo such slow transformation, however, (much slower than radium) that for all practical purposes they may be regarded as stable elements.

Small wonder, then, that the search for inactive representatives in Nature was abortive. Moreover, radioactive atoms can only occur in Nature if, like radium, they are generated continuously from elements of very long life, e.g., uranium. Were this not so, radium, if it had formerly been in existence, would have "died out" long ago.

2. ELEMENT 43: TECHNETIUM (Tc)

In 1925 NODDACK, TACKE and BERG¹ announced the discovery of an element 43, an *eka-manganese* to which they gave the

¹ W. NODDACK, IDA TACKE and O. BERG, *Naturwissenschaften*, 13 (1925) 567.

name *masurium* (after an ancient name for Prussia). Unlike that of rhenium (element 75), which was discovered by them at the same time, the existence of masurium has never been confirmed by isolation of the element. It may now, therefore, be taken as certain that the alleged discovery of element 43 in Nature was due to some mistake.

Representatives of element 43 arise out of β -radiating, artificially radio-active isotopes of element 42, i.e. molybdenum. Isotopes of the latter element can be obtained in various ways as, for example, by neutron capture in the isotopic mixture element molybdenum. These isotopes of molybdenum are also produced in appreciable quantities in the fission of uranium and can easily be extracted from the irradiated uranium after adding a few milligrams of a molybdenum salt. These molybdenum isotopes all radiate β -rays thereby becoming isotopes of element 43; two of these latter have definitely been proved to be transformation products of molybdenum isotopes. Three more are produced of which two appear to be very short-lived.

The first of the isotopes to be separated was a substance with 5.9 hours half-life which PERRIER and SEGRÈ extracted from 6.7 hour molybdenum. At the Kaiser Wilhelm Institute for Chemistry (HAHN and STRASSMANN, SEELMANN-EGGEBERT) the other isotopes were then found or their existence shown to be very probable.

Chart 1a shows the reactions together with their half-periods. Although we are merely dealing with quite

unweighable quantities, the mass-numbers can be taken as established with the possible exception of the mass-number 102 given in brackets.

We cannot here enter into details of the methods which often make it possible to deduce the mass-numbers of the various components taking part in a controlled nuclear reaction. Roughly, the method consists in irradiating an element, and also its neighbouring elements, with various particles, such as neutrons, protons and deuterons, to find out the properties of the atomic nuclei resulting from these controlled reactions and determine which of these reactions give rise to identical products. The various ways in which they are produced often enables one to deduce the atomic weight.¹

Chart 1a

ELEMENT 43: UNWEIGHABLE AMOUNTS OBTAINED BY NUCLEAR FISSION
(PRIOR TO THE URANIUM PILE)

Atomic weight	Mo 42	Tc 43	Ru 44	Rh 45	Pd 46
99	67 h →	5.9 h →	stable		
101	14.6 min →	14 min →	stable		
(102)	12 min →	< 1 min →	stable		
105*	5 min →	15 min →	4.5 h →	36.5 h →	stable
107		< 1.5 min →	4 min →	24 min →	?

* The half-lives 5 min and 15 min for atomic weight 105 are from SEELMANN-EGGEBERT. In the American report (Plutonium Project, already cited) they are only given as "short-lived" without further details.

¹ For a description how this is done for fission products of uranium, by comparing them with active isotopes produced by the reactions previously employed, see: W. SEELMANN-EGGEBERT and F. STRASSMANN, *Z. Naturforsch.*, 29a (1947) 80.

PERRIER and SEGRÈ were the first to supply specific information regarding the chemical properties of the 5.9-hour isotope of element 43. According to these investigators, the resemblance to the higher homologue rhenium is greater than that to the lower homologue manganese. Thus element 43 is precipitated both from dilute and from up to 5-normal hydrochloric acid by hydrogen sulphide; for higher acid concentrations the precipitation is no longer complete—in this respect it differs from rhenium. As in the case of rhenium the sulphide of element 43 is oxidized by hydrogen peroxide in alkaline solution into soluble anions corresponding to the perrhenate. On adding a little sodium perrhenate as carrier to this solution, element 43 may be quantitatively precipitated by nitron (general test for nitrate and perrhenate ions). We employed this reaction to separate element 43 from molybdenum and other fission products of uranium where these had not already been removed.

Subsequent investigation in the U.S.A. showed that the 5.9-hour isotope is what is termed an isomer to a much more stable representative of element 43. This more stable isotope could not be detected with the usual sources of radiation—e.g. in Europe. In the uranium pile, on the other hand, it is produced with yield thousands of million times as great. It is very long-lived, the half-life assigned to it being 4×10^6 years.

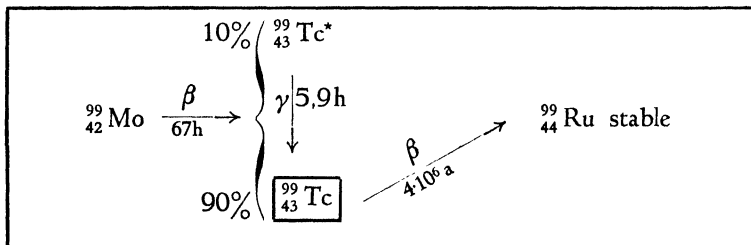
Ten per cent of the isotope of element 43 which is derived from the radio-active molybdenum isotope 99, is formed as an "isomer", i.e. in an excited state of 5.9 hours half-life,

it decays into the long-lived isotope. 90% of this isotope is produced directly. This isotope and also the other shorter-lived isotopes of Chart 1a emit β -rays and are gradually transformed into ruthenium, the next-higher element.

The chain of reactions, now definitely established, which give rise to the important isotope $^{99}_{43}\text{Tc}$ is shown in Chart 1b.

Chart 1b

ELEMENT 43: TECHNETIUM, SYMBOL Tc. WEIGHABLE QUANTITIES OBTAINED IN THE URANIUM PILE



Now that the existence of this long-lived representative of element 43 and its genesis in uranium fission have been established, a possibility is afforded of extracting the substance in quantities of use to the ordinary chemist. Actually, having a half-life period of over four million years, it is some thousands of times more stable than radium and thus behaves practically as a normal element.

Since PERRIER and SEGRÈ were the first to detect an isotope of element 43, it was left to them to choose a name for this element. The name they chose was "*technetium*", from

the Greek word "technetos", meaning "artificial", and the symbol is Tc.

Rough estimate of the possible output of technetium

By somewhat simplifying the reactions that take place in the uranium pile it is possible in the following way to arrive at a rough quantitative estimate. From the American reports it will be seen that for a pile run on pure graphite and pure uranium with the arrangement adopted in the U.S.A., for one part by weight of plutonium produced there will be roughly one part by weight of fission products. Let us assume that the fission reactions give 70 different products which sooner or later turn into stable isotopes contained in the Periodic Table; let us say 70 isotopes belonging, in round numbers, to 30 different chemical elements. To be exact one would have to take into account 85 reaction products with atomic weights from 72 to 158 and belonging to 34 different elements. The lowest and the highest atomic weights, however, are generated in such small amounts that they may be disregarded in our rough calculation.

Since an atom bomb consisting of plutonium must weigh at least 10 kg, it follows that during the formation of the 10 kg of plutonium in the pile, according to our assumption, formation of 10 kg of fission products or of their inactive end-products must have taken place, the "ashes", as it were, of the process. For the sake of simplicity we will assume that

equal amounts of fission products are formed (this is certainly not the case); this would give us $\frac{10,000}{70}$ g or, in round numbers, 140 g per plutonium bomb of 10 kg! More than 10 such plutonium bombs have almost certainly been made and therefore it follows that more than 1 kg of the new element technetium must have been produced in the uranium piles.

We may expect in due course of time that the new element 43 will be extracted at least by the gram so that its properties may be studied in much greater detail than was possible for PERRIER and SEGRÈ when they applied radioactive methods to the investigation of the short-lived 5.9-hour isomer.

3. ELEMENT 61: PROMETHIUM (Pm)

For element 61, the hitherto missing "rare earth", prospects are not quite so favourable as for element 43.

In a paper by SEABORG¹ entitled "Table of isotopes", not less than five artificial active isotopes of element 61 were listed—all of them as probable, not as certain. They were prepared by a great variety of irradiation processes, for the most part from neodymium, the next lower element. Authoritative information and atomic weight figures, however, found their first publication in the detailed report² on the fission products of uranium under the heading "Plutonium Project". The total number of isotopes listed is again five.

¹ G. T. SEABORG, *Rev. modern Physics*, 16 (1944) 1.

² *J. Am. Chem. Soc.*, 68 (1946) 2411.

In the case of two of these, not only has it been definitely established that they belong to element 61, but the atomic weights are also known. These isotopes are marked with the letter A in Chart 2. In the case of two others, while it has been established that they belong to element 61, the atomic weights assigned are only probably correct (marked B). The fifth is not certain and is marked D.

Chart 2 gives a summary of the data on element 61. The atomic weight of isotope 147 was also established by mass-spectrographic methods. The 55-hour isotope of element 61 with atomic weight 149 is given in the "Plutonium Project" list with 49 hours. The more recent value, however, is 55 hours.¹

Chart 2

ELEMENT 61: ALL ISOTOPES OBTAINED IN THE URANIUM PILE BY FISSION

Atomic weight	Nd 60	Pm 61	Sm 62	Eu 63	Gd 64	Degree of certainty
147	11.0 d →	3.7 a →	stable			A
149	(1.7h) →	55 h →	stable			B
151	(short) →	12 min →	20 h →	stable		D
153		< 5 min →	47 h →	stable		A
156		< 5 min →	10 h →	15.4 d →	stable	B

¹ M. C. INGRAM, D. C. HESS, R. J. HAYDEN and G. W. PARKER, *Physic. Rev.*, 71 (1947) 143.

Without the tremendous possibilities of the uranium pile it would have been a very difficult matter to establish a positive proof in the case of element 61—how difficult may be judged from the fact that of the five isotopes of element 61 announced in the *Reviews of Modern Physics* in 1944, not a single one has been included in the new Plutonium Project table. What had been announced as isotopes were evidently not isotopes of element 61. Of special interest to us is the isotope with atomic weight 147, this being the most stable. With its half-life of 3.7 years, this substance undergoes transformation about 400 times as rapidly as does radium (1600 years half-life); it is nevertheless 10 times as stable as polonium, the longest-lived isotope of element 84.

If we wish to form some idea of the possibilities of quantity production for this representative of element 61, we can adopt the same procedure as in the case of element 43. Let us take, for instance, a uranium pile that produces 10 kg of plutonium in the course of a few months; in the same period it will produce very roughly 10 kg of fission products, one of which will be the 3.7-year substance under consideration. Assuming for simplicity that each of the 70 (in round numbers) reaction series gives the same quantitative yield, it follows that the yield of the 3.7-years substance will be $\frac{1}{70}$ of the total amount—that is, as in the case of element 43, more than 100 g.

Such a calculation of the obtainable weight of a fission product, however, can only be correct if the period of irradiation

tion is short when compared with the half-life of the resulting substance. As we shall see later on, a formation period of a few months in the case of the very stable element plutonium is very short when compared with its half-life. And the same is also true for the 3.7-year isotope of element 61. The amount which accumulates is roughly proportional to the time of irradiation. This would not be true for the 55-hour isotope of element 61, because in a period of 10 half-lives, i.e. $10 \times 55 \text{ hours} = 23 \text{ days}$, the amount produced will be 99.9% of the equilibrium amount and even with continued production in the pile there will be no further accumulation of this substance, seeing that just as much will decay into samarium as is created from the neodymium.

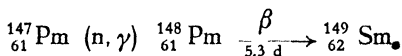
Long-term irradiation can therefore serve no purpose when the substance produced is shortlived. Once "radioactive equilibrium" has been reached, the increase in quantity comes to an end.

There is every reason to expect, therefore, that once the individual fission products have been separated out by various processes, the element 61 will be available by the gram or, if necessary, by the hundred grams. Weight for weight, i.e. in the ratio of 1600 years to 3.7 years, it would have an activity several hundred times as great as that of radium. On account of the intense radiation, work with appreciable quantities would involve considerable hazards.

All the same a further isotope of element 61 was obtained ¹

¹ G. W. PARKER, P. M. LANTZ, M. G. INGRAM, D. C. HESS and R. J. HAYDEN, *Physic. Rev.*, 72 (1947) 85.

by irradiation of the 3.7-year substance with neutrons leading to neutron capture:



This isotope 148 may therefore be regarded as a doubly-artificial product.

No details are yet available regarding the chemical properties of this new rare-earth.

Long before, many research workers had been trying to find this missing element from the series. In 1926, HOPKINS, YNTEMA and HARRIS, of the University of Chicago, thought, on the evidence of absorption and X-ray spectra, that they had found this element¹. They called it *illinium* after the State Illinois, shortly after which ROLLA and FERNANDES claimed to have discovered the same element², to which they gave the name *florentium*. Notwithstanding extensive research, the presence of this element in Nature has never been convincingly proved, which fact, in conjunction with the now discovered radioactivity of element 61, makes it fairly certain that it does not occur naturally.

Recently, the name *promethium*, with the symbol Pm, derived from the mythological figure Prometheus, was adopted for this element.

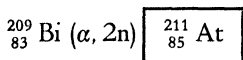
¹ J. A. HARRIS, L. F. YNTEMA and B. S. HOPKINS, *Nature*, 117 (1926) 792.

² L. ROLLA and L. FERNANDES, *Accad. Lincei, Atta 4* (1926) 498.

4. ELEMENT 85: ASTATINE (At)

We come now to element 85, an *eka-iodine*. All present there is no prospect of this element being produced in weighable quantities. We've already pointed out that two conditions must be satisfied before we can hope to get an artificial isotope in appreciable quantities: the substance must be a product of the chain reaction in uranium, and its half-life must not be too short. Neither of these conditions is satisfied by element 85. Actually, the isotope 211 was produced by CORSON, MCKENZIE and SEGRÈ¹ by bombarding bismuth with 32 million e.v. α -particles—that is to say, with particles of a type hitherto produced only in the LAWRENCE cyclotron. (The most penetrating α -particles of the natural radioelements RaC' and ThC' have energies of 7.6 and 8.6 million e.v. These are insufficient to overcome the repulsive forces of elements with high nuclear charges.)

The first reaction that takes place is:



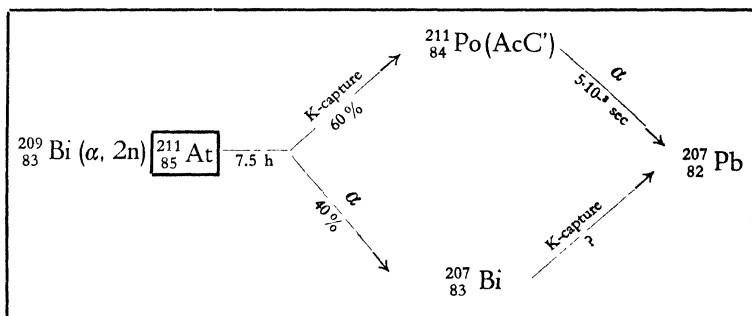
This product has a half-life of 7.5 hours and disintegrates in two different ways. About 60% is turned into polonium of mass 211, through K-electron capture by the nucleus from the electron-shell; the positive nuclear charge is in this way reduced by one unit. The product is the short-lived AcC'.

¹ D. R. CORSON, K. R. MACKENZIE and E. SEGRÈ, *Physic. Rev.*, 57 (1940) 1037; 58 (1940) 672.

which occurs naturally in uranium minerals, and turns into actinium-lead by emitting α -rays. The other 40% of the isotope emits α -rays and turns into a bismuth of mass 207, and this, in its turn, in all probability also changes into actinium-lead.

The series of reactions will therefore have the following form:

Chart 3
ELEMENT 85: ASTATINE (At)



The process of K-capture represents the incorporation of an electron from the innermost electronic shell ("K-shell") into the nucleus, with consequent neutralisation of one unit positive charge, i.e. transformation of the nucleus into one of the next lower element.

SEGRÈ and his co-workers proposed the name *astatine* for this eka-iodine isotope which they were the first to produce; the name comes from the Greek "astator", meaning "unstable", and was chosen by analogy with the English names for the

halogens—chlorine, bromine, iodine. (Since all the elements of nuclear charge 84 and over are unstable, one might well argue that the choice of name was not a very happy one.)

Chemically, of course, this eka-iodine, as a halogen, resembles its lower homologues in certain respects; in many reactions, however, it differs more widely from the halogens than the latter do among themselves. One surprising example of this difference is the fact that it is precipitated by hydrogen sulphide even from a strongly acid solution; another example is afforded by its electro-chemical deposition on zinc in sulphuric acid solution. In the latter respect it rather closely resembles polonium, its left-hand neighbour in the Periodic System.

Like the halogens, astatine can be precipitated with carbon tetrachloride, but after reduction with sulphur dioxide and acidification with nitric acid it is only incompletely precipitated by silver ions. When distilled with iodine, astatine is also distilled over—not so completely as the iodine but behaving, in this respect, quite differently from polonium.

Summarizing, we may say that eka-iodine occupies a kind of intermediate position between its next lower homologues and its left-hand neighbour in the Periodic Table, polonium.

In addition to this isotope of element 85 produced from bismuth, another artificial isotope has been detected. This new isotope was found as a member of a new radioactive transformation series originating in the element plutonium, which will be discussed later on. The new isotope

has a mass 217 and is an α -emitter of 2×10^{-2} sec half-life. The position of this eka-iodine isotope in the new transformation series will be indicated when we come to the element neptunium (p. 115).

Isotopes of eka-iodine (astatine) occurring naturally

Strictly speaking, we should confine our attention to the preparation and properties of the artificial new elements and leave the natural ones to be located elsewhere. We will, however, briefly refer to the representatives of element 85 which occur in natural uranium and thorium minerals and were detected by KARLIK and BERNERT of the Vienna Radium-Institute¹. These investigators have ascertained by painstaking experiments, that isotopes of element 85 are produced as by-products when what are termed the A-members of the three natural disintegration series undergo transformation. When these polonium isotopes disintegrate, it appears that side by side with the normal α -transformation a small fraction undergoes β -decay, giving rise to an isotope of element 85 which in turn emits an α -particle and becomes the C member of the main disintegration scheme, as indicated by the following diagram:

¹ B. KARLIK and T. BERNERT, *Z. Physik*, 123 (1944) 51; *Naturwissenschaften*, 31 (1943) 298; 33 (1946) 23.

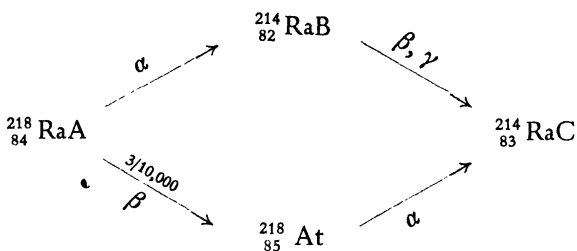


Fig. 6. Element 85 interlinking the A and C series of radioactive decay

The number of atoms decaying via the At isotope to that decaying via RaB bears the ratio $3/10,000$ for RaA, $1/10,000$ for ThA, and an even smaller value for AcA.

The range of the α -particles as measured by means of a 4 stage valve electrometer corresponds to the expected values for mass-numbers 218, 216 and 215 resp. in the cases of RaA, ThA and AcA resp.

It was only after the American scientists had succeeded in preparing the artificial active isotope of element 85 that KARLIK and BERNERT carried out their investigations. The right to christen element 85 therefore belonged to the Americans. Nevertheless it is to KARLIK and BERNERT that we must assign the honour of being the first to prove the existence of this naturally occurring isotope of the element and to establish its mode of formation.

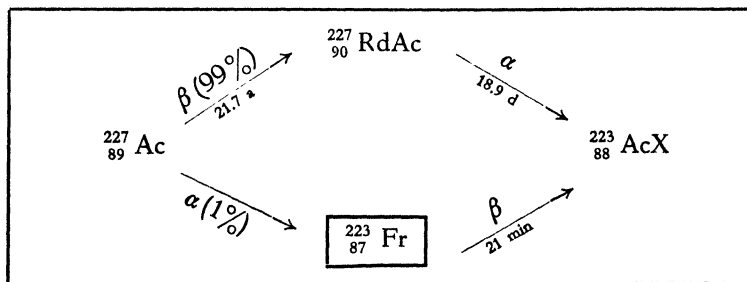
5. ELEMENT 87: FRANCIUM (Fr)

The position regarding element 87 is similar to that regarding element 85. So far there appears to be no prospect of its being

produced in weighable quantities. Until quite recently, in fact, the only isotope known was one existing in Nature and therefore outside our subject, which is restricted to artificial elements. For the sake of completeness, however, and because of the importance as the sole known representative of element 87, *eka-caesium*, it ought not to be missing from our list. It was detected by PEREY¹ as a branch product in the actinium decay series quite a number of years ago. Whereas actinium (element 89) normally emits β -rays and turns into the thorium isotope radioactinium, about 1% of the actinium emits α -rays and turns into an isotope of element 87. (On emission of α -rays the nuclear charge drops by 2 units and the mass by 4.)

The element is a β -emitter with a half-life of 21 min. It gives rise to AcX, a substance which has long been known and which is also formed out of the α -emitting radioactinium. The reaction is represented by the following diagram:

Chart 4
ELEMENT 87: FRANCIUM, Fr



¹ M. PEREY, *J. phys. Radium*, 10 (1939) 435, 439.

The properties of element 87 are such as would be expected of a higher alkali-metal. The name provisionally chosen by Miss PEREY was actinium K; now, however, the discoverer has proposed for this isotope of element 87—until quite recently its only known isotope—the name of *francium*, symbol Fr¹. From its origin we deduce that its mass is 223.

This representative of element 87 has now been joined by a further isotope—a short-lived member of the new radioactive transformation series of neptunium which we have already mentioned in connection with element 85. It has a half-life of about 5 min, its mass is 221 and it emits α -particles and is transformed into the short-lived astatine-isotope of mass 217.

6. ELEMENT 93: NEPTUNIUM (Np)

a. *Unweighable quantities*

We have already mentioned how certain elements investigated by FERMI, followed by HAHN, MEITNER and STRASSMANN and regarded by them as transuraniums, after the discovery of uranium fission turned out to be artificially active isotopes of elements of lower atomic numbers. What was known at the time about heavy nuclei quite justified, nay compelled the conclusion that we were dealing with transuraniums; and it was only when the fission of uranium was found to produce barium that the whole “transuranium” edifice began to totter and then came tumbling down.

¹ Cf. F. PANETH, *Nature*, 159 (1947) 8.

There is no doubt, however, that one of the reactions involved did, in fact, result in the formation of a representative of element 93. This was the β -ray transformation of the, already mentioned artificially active uranium-isotope, with a half-life of 23 min; this substance was definitely established by HAHN, MEITNER and STRASSMANN as an isotope of uranium using reliable chemical methods¹. It was not possible to identify, in an entirely satisfactory manner, FERMI's other short-lived β -emitters, which were taken to be uranium-isotopes leading to transuraniums, the difficulty being caused by the fact that they had half-lives of less than sixty sec; in the period before uranium fission, however, FERMI's verdict that they were uranium-isotopes appeared unassailable and was consequently accepted by us.

As a β -emitter, therefore, our 23-min isotope had to turn into a representative of element 93. Our search for this transformation product, however, proved fruitless. Our sources of radiation, which at the time consisted only of Ra-Be capillaries, were too weak. The substance in question was then identified in the U.S.A. by McMILLAN and ABELSON² as a β -emitter with half-life 2.3 days, the β -radiation at the same time indicating that this, the first of the transuraniums, must give rise to yet another transuranium, i.e. an isotope of element 94. The latter substance was not found to emit

¹ L. MEITNER and O. HAHN, *Naturwissenschaften*, 24 (1936) 158; O. HAHN, L. MEITNER and F. STRASSMANN, *Ber.*, 69 (1936) 905; 70 (1937) 1374.

² E. McMILLAN and T. H. ABELSON, *Physic. Rev.*, 57 (1940) 1185.

any observable radiation; it was evidently very long-lived, since, weight for weight, the radiation is inversely proportioned to the half-life. (The radiation from uranium is one three-millionth of that from an equal weight of radium; its half-life is therefore 3 million times as long).

The 2.3-day substance was submitted by McMILLAN and ABELSON to the usual radio-chemical methods for analysis of unweighable traces of a substance, with the object of discovering its chemical properties. It was found to exhibit a close resemblance to uranium, which gave strong support to the theory, already discussed by NIELS BOHR, V. M. GOLDSCHMIDT and others, that in the neighbourhood of uranium there are elements resembling one another in properties and forming a group comparable to that of the rare-earth elements. Element 93, however, differed notably from uranium in the readiness with which it passed into the tetravalent state and it was this difference in behaviour that rendered it possible to separate it from uranium.

As already related, element 93¹ was then christened by the discoverers of the 2.3-day isotope with the name of *neptunium* (symbol Np) after the planet Neptune which is the next planet beyond the planet Uranium.

After this STARKE¹ succeeded in separating the 2.3-day body by a method different from the one followed by the American scientists, and HAHN and STRASSMANN², using a

¹ K. STARKE, *Naturwissenschaften*, 30 (1942) 107.

² O. HAHN and F. STRASSMANN, *Naturwissenschaften*, 30 (1942) 256.

modest high-voltage plant for the purpose of preparing stronger samples, without difficulty produced the 2.3-day body and, after establishing quite a number of its properties, worked out in detail a separation procedure by which this isotope of element 93 can be prepared in a radioactively pure state freed not only from the hexavalent uranium but also from the tetravalent thorium and the numerous fission products.

This separation-procedure is roughly as follows: The uranium solution containing element 93 and the numerous fission products is heated for a short time with sulphur dioxide; this brings element 93 into the tetravalent state, the uranium remaining hexavalent. This is followed by separation of the bulk of the uranium as uranyl-sodium-acetate free from element 93. In the filtrate the slight uranium residue together with element 93, uranium X and a part of the fission products are separated from the numerous neutral salts by precipitation with ammonia, then dissolved in nitric acid, after which element 93 is brought into the hexavalent state by means of potassium bromate. Uranium and element 93 are now separated from the residual fission products and the bulk of uranium X by the almost complete removal of the uranium by means of sodium acetate. The slight quantities of uranium and uranium X are precipitated from dilute acetic acid solution by 8-hydroxyquinoline (oxin). The element 93 remains in bulk in solution—free from uranium, uranium X and the fission products. The oxin solution is kept slightly ammoniacal with ammonia and is freed from oxin by prolonged boiling. There must be complete removal of oxin, as otherwise element 93 will not separate out in bulk. Element 93 can now be separated quantitatively from the oxin-free solution by ammonia together with a few mg of iron, zirconium or the like to act as carriers.

1940 saw the discovery of yet another isotope of element 93.¹ This isotope was produced in the great LAWRENCE cyclotron by irradiating uranium with deuterons. The reaction involves absorption of a deuteron and liberation of two neutrons with formation of an isotope of element 93 having the mass of the irradiated uranium 238. This new substance has a half-life of 2 days, emits β -rays and becomes transformed, like the 2.3-day isotope of mass 239, into an isotope of element 94. In this case it was also soon established that element 94 is an α -emitter with a half-life of about 92 years which disintegrates into uranium 234 – the isotope that forms such a minute proportion of the mixed element uranium.

In the meantime Japanese scientists² had established yet another transformation process in uranium which also led to a representative of element 93. A very fast neutron impinging on uranium is not absorbed but, on the contrary, knocks out another neutron. In this case uranium 238 is not transformed into a uranium-isotope 239 as in the above-mentioned case of absorption, but into one of mass 237. The latter, like the 23-min uranium, is also a β -emitter – 6.8 days half-life – and must also turn into the next higher element, i.e., become an isotope of neptunium, a long-lived isotope as otherwise it would have been detected immediately.

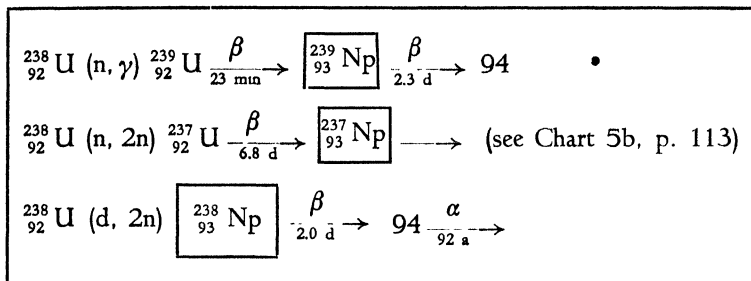
The various reactions, as they were known before the construction of the first uranium pile, are shown in Chart 5a :

¹ G. T. SEABORG, E. McMILLAN, A. C. WAHL and J. W. KENNEDY (1940), reported in "The Impact of Nuclear Chemistry", by G. T. SEABORG, *Chem. Eng. News*, 24 (1946) 1192.

² Y. NISHINA, T. YASAKI, K. KIMURA and M. IKAWA, *Phys. Rev.*, 58 (1940) 661.

Chart 5a

ELEMENT 93: NEPTUNIUM — UNWEIGHABLE (PREVIOUS TO URANIUM PILE)



b. Weighable quantities

We have already mentioned what happened in America when it became known that it was possible to utilize the chain-reaction as a source of energy. The decision was taken to concentrate all the resources of the State on the exploitation of nuclear energy for the production of super-explosives. One way of achieving this was by separating the requisite quantities of uranium 235 out of uranium 238, a very difficult and laborious separation, being possible only by isotope-separation methods. The other way was provided by the uranium pile and consisted in producing the evidently long-lived transformation product of the 2.3-day neptunium, viz. the 239-isotope, unknown at that time, of element 94. It was to be expected that in this substance of odd mass-number fission could be induced by neutrons of all energies, just as in the

case of uranium 235, so that it could be used in a similar way as an explosive. The concentrated efforts which were made to produce element 94 will be considered in greater detail when we come to deal with that element later on (p. 119).

In the fission processes of the uranium pile the neutrons which are constantly liberated are, at the outset, particles of high energy. As we have seen, they have the power of knocking out another neutron from uranium 238 and forming the above-mentioned 6.8-day isotope of uranium. This reaction is a less frequent one compared with the capture of a neutron by the 23-min uranium of mass 239; nevertheless, owing to the tremendous increase in yield during the formation of artificial isotopes in the pile, it sufficed to establish the neptunium isotope of mass 237, undetected before the advent of the pile, as a very stable substance with a half-life of 2.25×10^6 years and to permit of its being separated and submitted to a thorough examination. This body emits α -rays and does not, therefore, form the next higher element on disintegration but turns into an isotope of element 91, protactinium, with mass-number 233. (This last-mentioned isotope had already been produced, before the fission of uranium, by MEITNER, STRASSMANN and myself, by irradiating thorium with slow neutrons ¹.)

The α -emitting neptunium-isotope of mass 237, with its

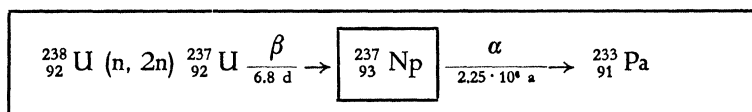
¹ L. MEITNER, F. STRASSMANN and O. HAHN, *Z. Physik*, 109 (1938) 538; O. HAHN and F. STRASSMANN, *Naturwissenschaften*, 29 (1941) 285.

half-life running into millions of years, is an element capable of being treated like any ordinary chemical element. Although the yield in a uranium pile is slight compared with that of elements which are more readily created by irradiation with slow neutrons, an amount weighing several 100 mg had already been produced in 1946.

The reaction-sequence resulting in the creation of this long-lived isotope of neptunium is shown in Chart 5b:

Chart 5b

NEPTUNIUM, WEIGHABLE (PRODUCED IN THE URANIUM PILE)



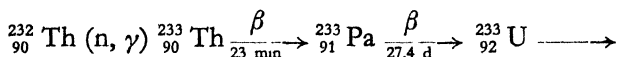
The properties of neptunium in unweighable quantities had been investigated by radio-chemical methods; they could now, of course, be much more thoroughly investigated with the weighable quantities supplied by the pile. A number of pure compounds were prepared. Analogous to the uranyl salts, for example, there are neptunyl salts, isomorphous with the corresponding uranium compounds; and in addition to the previously found tetravalence and hexavalence of the element it was also possible to produce pentavalent and trivalent compounds with a tendency showing preference towards formation of the tetravalent oxidation-stage.

c. New artificially-radioactive series ($4n + 1$)

The provision of a pure isotope of neptunium in weighable quantities has made it possible for us to follow the course of the transformation-products right down to the inactive end-product. Out of neptunium we get a long series of artificially active isotopes. As this series includes a representative of each of the new elements francium and astatine, we will consider it a little more closely.

The three great radioactive families, which have long been known to exist in nature in uranium and thorium minerals, have their origin in uranium 238 (*uranium series*), uranium 235 (*actinium series*) and thorium 232 (*thorium series*); their mass-numbers are $4n + 2$, $4n + 3$ and $4n$, where "n" is a whole number (e.g. for uranium 238 = $4 \times 59 + 2$; for thorium 4×58 etc.). There is no series $4n + 1$ known among natural radio-elements.

The first representatives of the missing $4n + 1$ series were discovered by MEITNER, STRASSMANN and myself. On capture of a neutron by thorium 232 a β -emitting thorium-isotope of mass 233 is created—i.e. a representative of the $4n + 1$ series¹ which then gives rise to a protactinium-isotope of the same mass². The successive reactions are as follows:



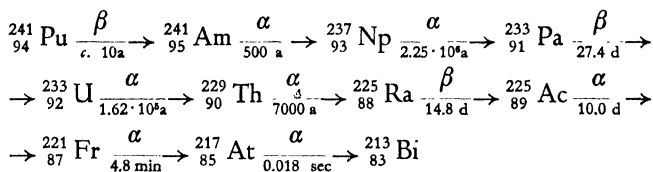
¹ O. HAHN and L. MEITNER, *Naturwissenschaften*, 23 (1935) 320.

² L. MEITNER, O. HAHN and F. STRASSMANN, *Z. Physik*, 109 (1938) 538.

With the weak radiation sources at our disposal we were unable to detect this isotope of uranium. With the tremendous neutron intensities of the pile, however, it was soon identified as a long-lived α -emitter (half-life = 1.62×10^5 years). Now the very same isotope is also produced out of neptunium, via the above-mentioned protactinium-isotope, the creation of which we again checked and confirmed after the discovery of uranium-fission¹.

The long-lived artificial uranium 233 also undergoes transformation and the new $4n + 1$ series may be taken as beginning with this uranium-isotope or better, perhaps, with the even more stable neptunium or the somewhat less stable plutonium.

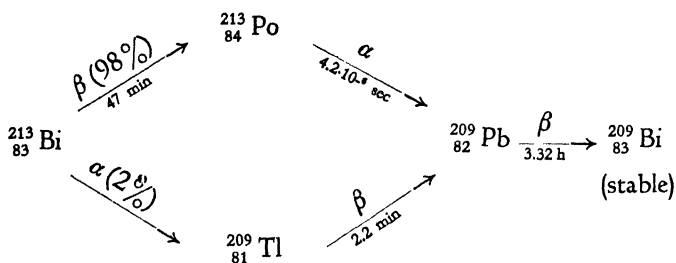
The series (usually called the *plutonium-neptunium* series) will then have the following form:



This series contains the elements francium and astatine which we have already mentioned, their half-lives being francium 4.8 min and astatine 2×10^{-2} sec. There is, therefore, no possibility of these elements being produced in weighable quantities.

The new $4n + 1$ series then continues as shown below:

¹ O. HAHN and F. STRASSMANN, *Naturwissenschaften*, 29 (1941) 285.



The elucidation of this long artificial transformation series occurred apparently simultaneously in the Argonne National Laboratory in Chicago¹ and in the Division of Atomic Energy, Chalk River, Ont., Canada² and in both cases the work was carried out by teams of chemists and physicists.

7. ELEMENT 94: PLUTONIUM (Pu)

Plutonium is, without doubt, the most important of all the artificially produced new chemical elements and an account of its first production in weighable quantities is not only very interesting generally, but is also very instructive for chemists.

After the discovery of the 2.3-day isotope of element 93 it was immediately concluded that there would have to be formation of a very stable isotope with atomic number 94 and atomic weight 239. For the 2.3-day substance emits β -rays and must consequently turn into element 94; and as

¹ F. HAGEMANN, L. I. KATZIN, M. H. STUDIER, A. GHIORSO and G. T. SEABORG, *Physic. Rev.*, 72 (1947) 252.

² A. C. ENGLISH, T. E. CRANSHAW, P. DEMERS, J. A. HARVEY, E. P. HINCKS, J. V. JELLEY and A. N. MAY, *Physic. Rev.*, 72 (1947) 253.

this element was not immediately detected, even among the very strong samples of the 2.3-day substance furnished with the help of the cyclotron, it followed that the looked for isotope was sending out very few rays, i.e. it would have to have a very long half-life. •

When discussing neptunium we mentioned that on irradiating uranium 238 with deuterons absorption of deuterons and liberation of two neutrons with formation of a neptunium-isotope takes place, which is then transformed into a representative of element 94 with the convenient half-life of 92 years. This half-life of 92 years on the one hand, with the sources available in the U.S.A. allowed production of the 238-isotope in sufficient quantities to render it easily detectable by means of its α -rays, and on the other hand its activity remained practically constant during the time it took to perform an experiment.

Of course the quantities involved were not yet of the weighable order, but, as in the case of the 2.3-day neptunium, it was also possible in this case to gain important information regarding the chemical character of element 94. In the same way as element 93, this element resembles uranium to the extent that it appears in the hexavalent and tetravalent states. The tendency towards tetravalence (and even towards trivalence), however, was more pronounced in element 94 than in element 93, and stronger oxidizing agents were required to get it to pass into the higher states of oxidation. Nevertheless, plutonium can also exist as the plutonyl ion and is in

this respect, like the neptunyl-compounds, isomorphous with the uranyl-compounds.

The scheme of nomenclature which had given the name neptunium to element 93 now led to the name *plutonium* for element 94. •

Following the detection of this fairly stable plutonium isotope, obtainable only with the aid of the cyclotron but not from the uranium pile, interest became centred around the as yet unknown long-lived transformation product of 2.3-day neptunium, of mass 239, and in 1941 the product in question was discovered at the University of California¹. It proved to be an α -emitter with a half-life of 24,000 years.

The tremendous importance of this plutonium-isotope was immediately grasped in the U.S.A. For all practical purposes it could be regarded as stable, but, like U235, fissionable by fast and by slow neutrons. When produced in sufficient quantity, therefore, it could be used as a super-explosive. Its separation from uranium could be effected by chemical methods and would therefore present a much simpler problem than the separation of uranium 235 from its isotope uranium 238.

American *physicists* now bent all their efforts to the task of erecting plant which would allow the formation, on the one hand, of adequate amounts of plutonium from the resonance

¹ G. T. SEABORG, E. SEGRÈ, J. W. KENNEDY and E. O. LAWRENCE (1941), reported in G. T. SEABORG, "The Impact of Nuclear Chemistry", *Chem. Eng. News*, 24 (1946) 1192.

capture by U238 of neutrons, and, on the other hand, of fission products of U235 in quantities sufficient to keep up a continuous chain-reaction. Their efforts culminated in the "uranium pile" (see p. 55).

It was the task of the *chemists* to devise methods for separating the plutonium from the uranium which it resembled and also from the fission products whose intense radioactivity made them dangerous to handle.

It was essential, of course, that the methods of separation should be tried out in detail in order that a proper basis might be provided for planning and erecting the enormous plant that would be required for the production of plutonium on a large scale. The attempt was therefore made, by means of the biggest cyclotrons available in the U.S.A., to provide plutonium not only in sufficient quantity to permit of its being studied by radio-chemical tracer methods, but in quantities which, though small, would be weighable and thus provide guidance of greater reliability. Hundreds of pounds of uranium were submitted to neutron-bombardment for several months and success was achieved first in the formation of a few thousandths of a milligram and later of a whole milligram of plutonium.

Every available means had been brought into service and for the very first time a chemical element had been artificially produced in weighable quantities. Newly developed "ultra-microchemical" methods of investigation were then brought to bear upon thousandths of a milligram of the product and

methods of separation of such reliability were worked out from these minute quantities, that when the uranium piles were ready to deliver the material, the huge chemical plant had already been erected and stood ready to extract the plutonium by the kilogram.

These ultra-microchemical methods are described in detail, with illustrations of the special apparatus, in SEABORG's report: "*The Impact of Nuclear Chemistry*", to which we have already referred¹. Test-tubes and beakers of from 0.1 to 0.00001 cm³ capacity, in the form of very fine capillaries, were employed together with pipettes and micro-manipulators of corresponding dimensions. A description is given of specially constructed micro-balances which can weigh 1 microgram with an accuracy of 0.02 microgram, at a maximum load of 25 mg. Investigations were carried out on solid precipitates separated from solution by centrifuge and weighing from 10⁻⁷ to 10⁻⁵ g and even the pure metallic plutonium was itself prepared and its behaviour tested in these minute quantities.

In Chapter II, section 6, we have already mentioned the proper pride with which SEABORG points out in his report that the amounts involved in the ultra-microchemical experiments and the amounts later on separated by the huge plant which was based on those experiments, bore to one another the ratio of 1 : 10¹⁰—a wonderful achievement indeed on the part of the numerous scientists who were engaged on the task.

¹ Another recent publication on this subject is the monograph by P. L. KIRK, *Quantitative Ultramicroanalysis*, New York, 1950.

Quite a number of pure compounds of plutonium and even the metal itself were now prepared in fractions of a milligram with the result that very soon the chemistry of plutonium was "known as well as, or even better than, the chemistry of many an ordinary chemical element in the Periodic System". Compounds were prepared with plutonium in the 6-, 5-, 4- and 3-valent states and, as we have already mentioned, it was found that the preferred state of the element was the tetravalent one.

Now that it is possible, in the uranium-pile, to produce plutonium in greater quantity, there is no longer any necessity to work with microscopic amounts. Normal chemical methods may be applied and there is no doubt that the initiated are fully conversant with all the properties both of the element itself and of its compounds. Solutions of the element vary in colour according to the state of valence. Solutions of 4-valent compounds are generally orange or brown (with the exception of the nitrate, the solution of which is green). Trivalent compounds are a brilliant blue. In valences 3, 4 and 6, the properties resemble those of 3-valent lanthanum, 4-valent thorium and 6-valent uranium.

The quantitative analysis is generally carried out with the 4-valent compounds in the form, for example, of $\text{Pu}(\text{SO}_4)_2$, PuO_2 , $\text{Pu}(\text{NO}_3)_4$. The determination can, however, also be effected by radioactive methods. From the half-life of 24,000 years we get that the number of α -particles emitted per milligram of Pu per minute is 1.6×10^8 . The Pu-content

may therefore be found directly by counting the α -particles in a thin layer suitable for this purpose¹.

Since 1945 the production of plutonium in the uranium piles has certainly amounted to hundreds of kilograms. Owing to its long half-life of 24,000 years small quantities of the element can be used in experimental work without any risk of radiation injury. On the other hand the long half-life means that even a microscopic amount of plutonium, if deposited in the body will continue to irradiate the neighbouring cells at a practically constant rate, and extreme precautions against inhalation and ingestion of plutonium dust have to be observed. It is only when the critical weight has been attained, i.e. that weight of plutonium at which more neutrons remain within the substance than escape through the surface, that we get the devastating explosive that has rocked the world.

The chain reaction leading to the explosion sets in as soon as the critical weight is reached. As stated on p. 60 this critical weight for plutonium has not yet been disclosed. Estimates fluctuate between 10 kg and an upper limit of 30 kg, a piece, therefore, of about the size of a skittle-ball, a coconut or even smaller.

Such pieces when divided into two halves would still be quite harmless. The explosion results when the two halves are brought together as speedily as possible, say by shooting

¹ H. J. EMBLÉUS, "*Some Aspects of Nuclear Chemistry*", The Royal Institute of Chemistry, London, 1947.

one half on to the other. No special means of ignition are required since there are always one or two neutrons about and a single one suffices to initiate the chain.

If the two parts are not brought together with great rapidity, the starting up of the reaction would lead to the vaporization of much of the bomb material and that portion would thus escape fission. To reduce such losses to a minimum it seems certain that the whole arrangement is securely encased in strong metal walls which scatter back the neutrons. Detailed information regarding this matter have not been published.

Let us assume (cf. Chapter II, section 9) that in an explosion only 20% of the material undergoes fission while the remaining 80% escapes fission owing to premature vaporization. From 10 kg of plutonium we then get 2 kg of a mixture of the most widely different, active isotopes of ordinary chemical elements!

At the moment of explosion and immediately thereafter, the radiation effects must assume almost inconceivable dimensions. The 2 kg of artificially active fission products are for the most part very short-lived substances, with half-lives in many cases amounting to only a few sec or min, and each one initiates a series of radioactive transformations¹. If we compare the activity of a substance with half-life 1 min with that of radium, which has a half-life of 1600 years, we find that 1 mg of the former gives the same intensity of radiation as $1600 \times 365 \times 24 \times 60$ mg of radium. This gives about

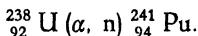
¹ See, e.g., the tables in the above cited publications of W. SEELMANN-EGGEBERT and F. STRASSMANN, O. HAHN and Plutonium Project.

800 kg of Ra for 1 mg of the substance, and in the 2 kg there are many such short-lived substances!

Owing to the short lives of most of the artificial fission products, however, the enormous activity rapidly diminishes. On the other hand it has also been established that some of the substances produced have half-lives measurable in days, months and years. In these cases there is only a gradual diminution in activity; weight for weight, however, this activity is proportionately smaller.

The only plutonium-isotopes hitherto mentioned, ^{238}Pu and ^{239}Pu , are both α -emitters; they consequently disintegrate into uranium-isotopes of mass 234 (long known, from natural radioactivity, as U II) and into U235.

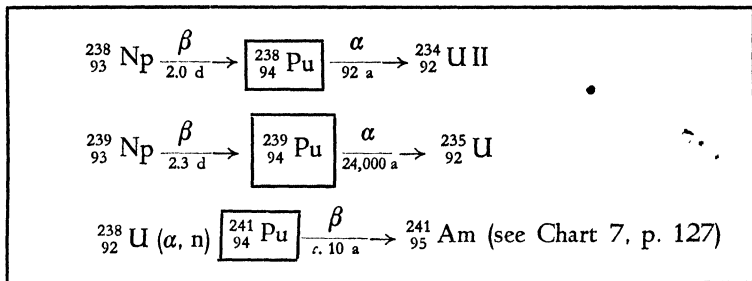
There is a third and very important plutonium-isotope which has been produced by SEABORG and co-workers¹ by bombarding uranium with the 40 million v α -rays of the Berkeley cyclotron. The uranium captures an α -particle and emits a neutron, the resulting product being a plutonium-isotope of mass 241:



This plutonium-isotope has a rather long life and emits β -rays. The resulting product must therefore be an isotope of the next-higher element 95. Before we proceed to discuss this new trans-uranic product, however, we will first of all summarize, in Chart 6, the reactions which lead to the formation of plutonium.

¹ G. T. SEABORG, *Chem. Eng. News*, 24 (1946) 1192.

Chart 6
ELEMENT 94: PLUTONIUM, Pu



Strangely enough, plutonium also seems to appear very scantily in Nature, the only example of a natural transuranium. SEABORG¹ reports having found traces of plutonium in pitchblende, in a ratio of 1 : 10¹⁴. No satisfactory explanations has yet been suggested as to how plutonium is formed from uranium by a natural process.

8. ELEMENT 95: AMERICIUM (Am)

It will be seen from Chart 6 that when uranium is bombarded with extremely fast α -particles, formation of a β -emitting plutonium of mass 241 takes place. Its transformation product is an isotope of element 95 with mass 241 which has a half-life of 500 years and undergoes α -decay into the long-lived, α -emitting element neptunium 237.

SEABORG gave element 95 the name of *americium* (for the explanation of this name see p. 139).

¹ G. T. SEABORG, *Chem. Eng. News*, 23 (1945) 2190.

This element is not produced in the uranium pile because it is not the product of a neutron-induced reaction. For the time being, therefore, we cannot expect it to be produced like plutonium and neptunium by the gram, much less by the kilogram.

From a report by SEABORG (see statement in a lecture in the Rochester Section of the American Chemical Society, of Nov. 18, 1946) it appears that americium, in the form of americium hydroxide $\text{Am}(\text{OH})_3$, has been obtained by CUNNINGHAM in quantities amounting to fractions of a milligram. The method was no doubt similar to that adopted in producing the first weighable quantities of plutonium: utilization of the most powerful cyclotrons for producing the strongest beams of α -particles, protracted irradiation of uranium for producing the β -emitting Pu-isotope, separation and preparation of the pure element 95 by the ultramicrochemical methods elaborated for dealing with plutonium.

There are, of course, limits to the thickness of the layers of uranium used for irradiation with the α -particles, since quite thin layers suffice to absorb even the 40 million e.v. α -particles. On the other hand the particles delivered by the cyclotron are available directly, whereas the neutrons have to be produced indirectly from the charged cyclotron-particles (generally deuterons) by means of a nuclear process.

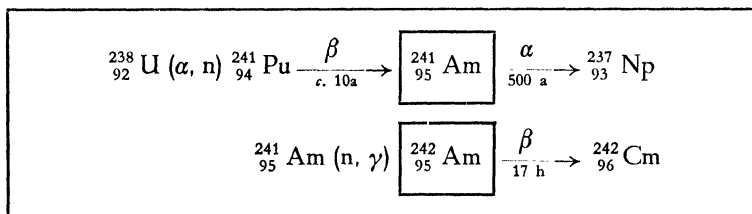
In the case of americium the tendency towards trivalence is even more pronounced than in the case of elements 93 and 94. The substance is still very inaccessible, however, and exact details are not yet available.

The usual neutron-capture process has been applied to $^{241}_{95}\text{Am}$ and has resulted in the formation of a further $^{242}_{95}\text{Am}$, which is a matter of considerable importance. And whereas the isotope 241 emits α -rays and degenerates into an element 93, the Am 242, like most active species of atoms produced by neutron-capture, emits β -rays and thus carries on the formation of transuranic elements.

The two reactions which lead to the formation of americium-isotopes are shown in Chart 7:

Chart 7

ELEMENT 95: AMERICIUM, Am



9. ELEMENT 96: CURIUM (Cm)

Neutron-capture by the relatively stable americium 241 produces the 18-hour americium-isotope of mass 242, which emits β -rays with formation of element 96, to which SEABORG has given the name of *curium* (explanation see p. 139).

Owing to its mode of formation from $^{242}_{95}\text{Am}$, this curium-isotope also has mass 242. It is an α -particle emitter with a half-life of 5 months and decays back into the plutonium 238.

Another way of preparing this representative of element 96

is by irradiating $^{239}_{94}\text{Pu}$ (the isotope which has been produced by the kilogram in the uranium pile) with the fastest α -rays of the Berkeley-cyclotron. This is an (α, n) -process, α -rays being absorbed and a neutron being liberated.

Astonishing enough, the same method also gives rise to yet another curium-isotope; in this case the α -rays liberate not one but three neutrons, the process therefore being an $(\alpha, 3n)$ -process. The new isotope has consequently to be assigned the mass 240.

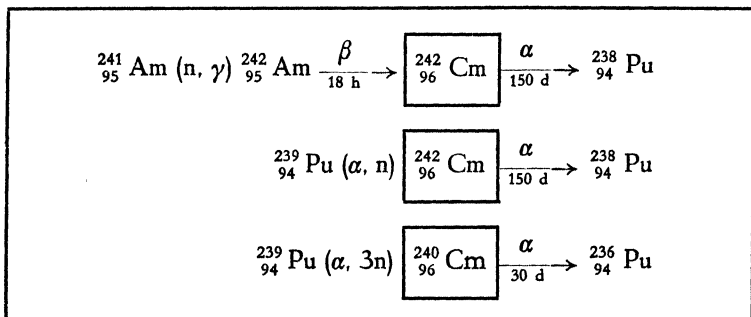
The half-life of this curium-isotope is only one month; like the 5-month isotope it also emits α -rays and appears to decay into a plutonium 236.

The reactions which lead to the formation of curium are summarized in Chart 8.

Owing to the fact that these isotopes of element 96 are obtained indirectly (via americium) or directly (from plu-

Chart 8

ELEMENT 96: CURIUM, Cm



tonium) by means of α -particle reactions, there is, at present, no prospect of element 96 being produced in weighable quantities; it is to the pile that we look for weighable quantities and the pile is not a source of intense α -rays.

The chemical properties of element 96 have therefore been established only by the methods of radio-chemistry from the activities observable when the substance is in a highly diluted state.

Even if the curium-isotopes which have so far been discovered were produced in weighable quantities, they would certainly be dangerous substances and very difficult to manipulate on account of their intense radioactivity. As compared with radium, for example, the more stable of the two isotopes would emit, weight for weight, over 3,000 times as many α -particles. Presumably there would also be strong emission of α -radiation at the same time. Concentrated solutions of the element would grow hot and decompose the water and, owing to radioactive recoil, the drops of a concentrated solution would not hold together but would be gradually sprayed away; it would indeed be difficult to work with such a substance.

The radio-chemical methods suffice, however, for determining the properties, just as they have had to suffice for most of the naturally radioactive elements and, until a few years ago, for all the artificially radioactive elements.

The trend which was clearly shown by elements 93, 94 and 95, is confirmed by the element curium; the increasing stability of the trivalent compounds, according to SEABORG,

becomes so pronounced in the case of curium as practically to exclude the occurrence of any but trivalent compounds. The element thus behaves exactly like a homologue of actinium and resembles the rare earths in many ways.

10. ELEMENT 97: BERKELIUM (Bk)

Very recently, THOMPSON, GHIORSO and SEABORG¹ have succeeded in obtaining a representative of element 97 by bombardment of $^{241}_{95}\text{Am}$ with 35-MeV α -particles in one of the huge Californian cyclotrons.

The element has been named after the town Berkeley, in which it was discovered, *berkelium*. The half-life is given as 4.6 hrs with a probable atomic weight 243. It decays by electron capture and with about 0.1 % α -particle-branching; there seem to be three different α -particle groups, the highest of which is 6.72 MeV².

The separation of berkelium from the bombarded americium was accomplished by precipitation and adsorption methods, in which use was made of the increasing trivalency in the direction of increasing atomic number.

11. ELEMENT 98: CALIFORNIUM (Cf)

The production of element 98 has just been reported by the

¹ S. G. THOMPSON, A. GHIORSO and G. T. SEABORG, *Phys. Rev.*, 77 (1950) 838.

² For reference also see F. PANETH, *Nature*, 165 (1950) 748.

same Californian team and was achieved in an analogous way as their production of Bk¹. By irradiating the isotope of element 96 (curium), of atomic weight 242, a new nuclide was formed which decays with a half-life about 45 min, emitting α -particles of energy 7.1 MeV. The possible and, for theoretical reasons, likely decay by electron capture had not yet been observed. The atomic weight should be 244. The element has been named after the state California, in which it was discovered, *californium*.

Of course, neither berkelium nor californium is obtainable in weighable quantities. The demonstration of their existence and their closer investigation could only be carried out with the help of "tracer" methods.

12. DECAY SERIES OF THE NATURAL AND ARTIFICIAL "ACTINIDES"

In Figures 7 and 8 (p. 132–135) the natural and artificial decay series ("families") as well as their mutual interlinkings have been collected. The natural series are the thorium series ($4n$), the uranium series ($4n + 2$), and the actinium series ($4n + 3$), whereas the plutonium series ($4n + 1$) and the protactinium branch of the uranium series ($4n + 2$) are artificial. The series themselves are constituted by α - and β -transmutations, the starting elements of the artificial series originate through (n, γ) - and (α, n) - processes.

¹ S.G. THOMPSON, K. STREET (Jr), A. GHIORSO and G. T. SEABORG, *Phys. Rev.*, 78 (1950) 298.
For reference see also F. PANETH, *Nature*, 165 (1950) 748.

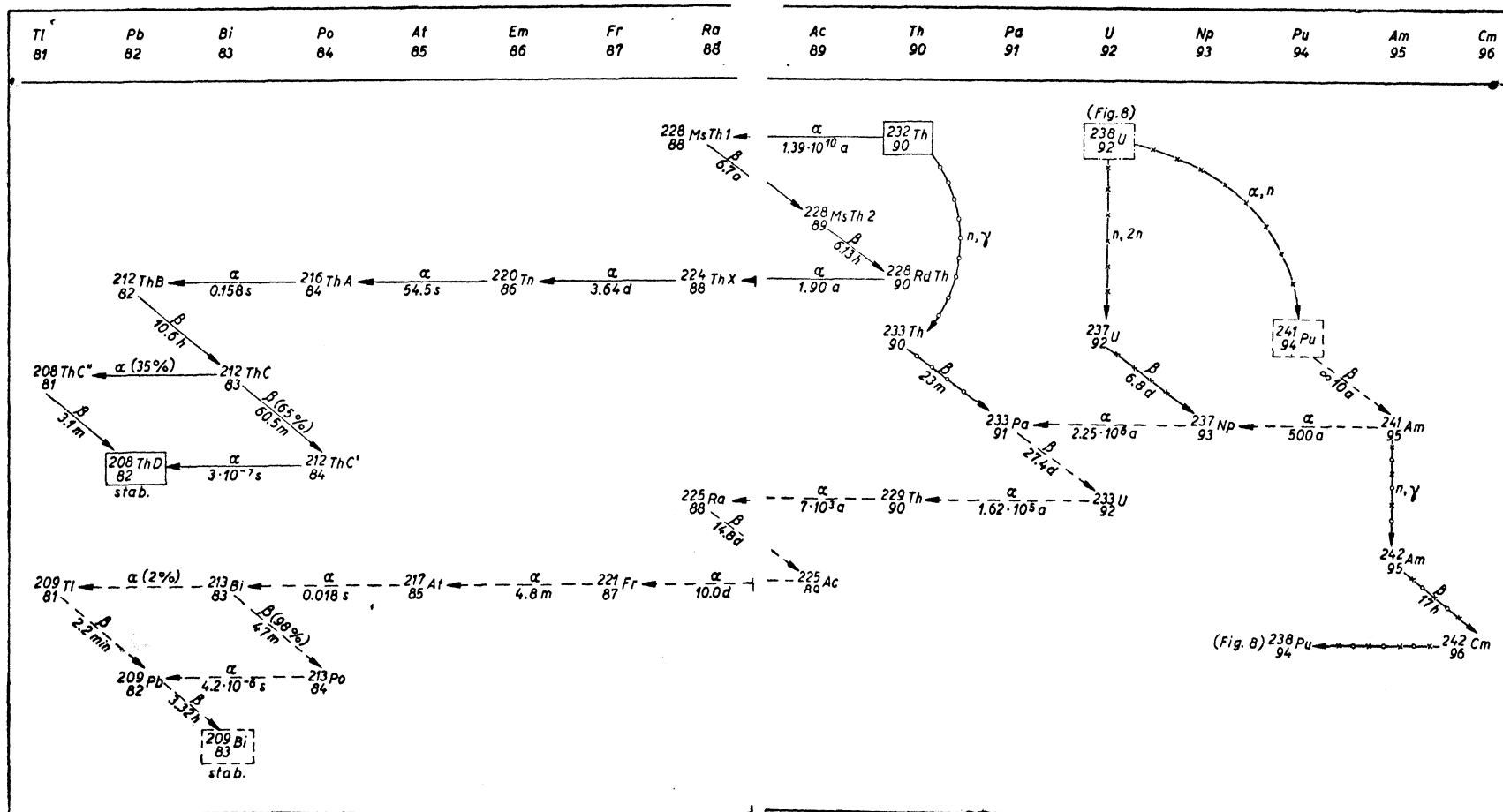


Fig. 7. Radioactive series (see also Fig. 8).

———— Thorium series 4n
 - - - - - Plutonium series 4n + 1

—○— Bridge from thorium to plutonium series
 —×— Bridge from uranium (Fig. 8) to plutonium series
 —○— Bridge from plutonium to uranium series (Fig. 8).

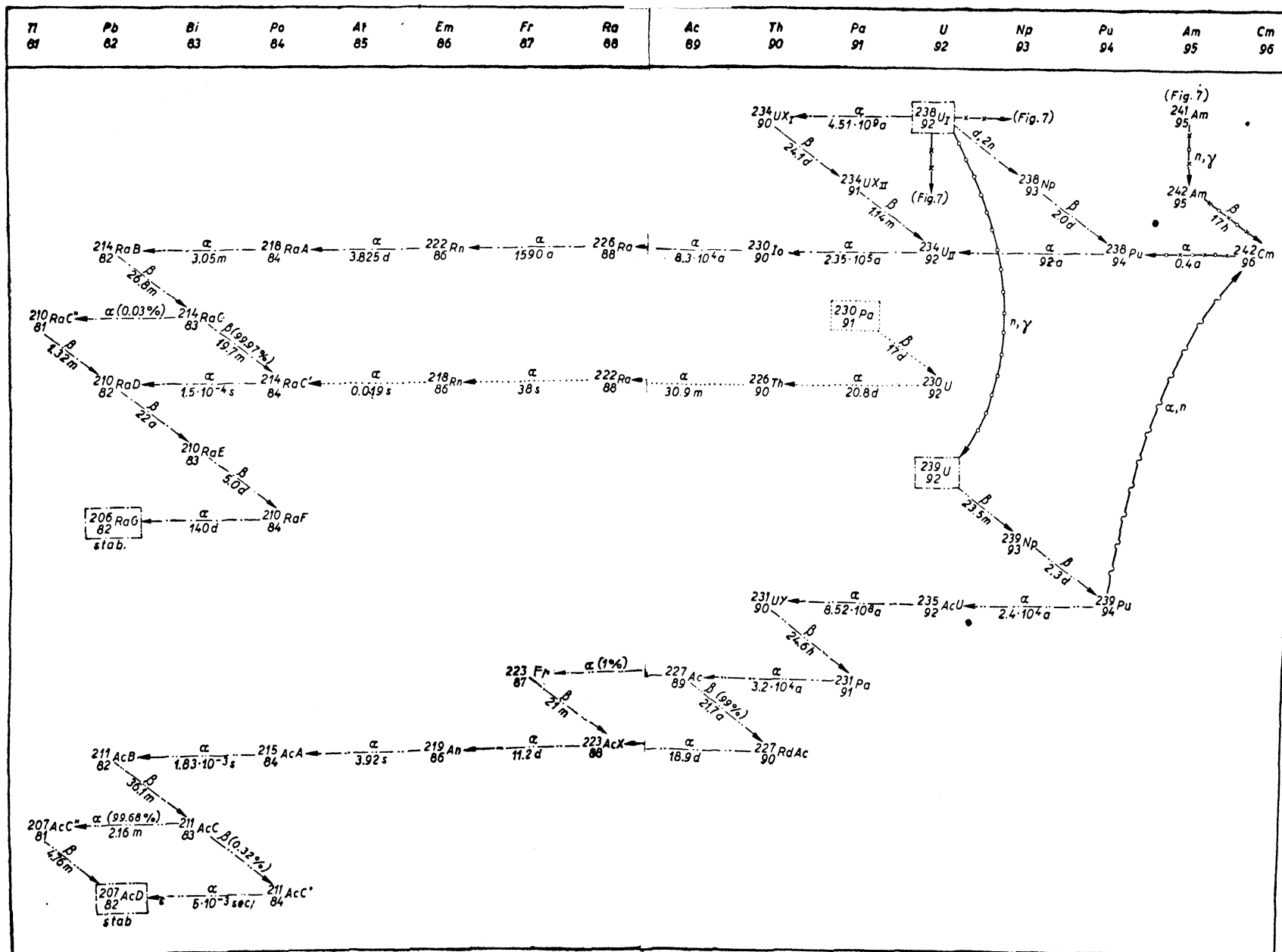


Fig. 8. Radioactive series (see also Fig. 7).

- Uranium series $4n + 2$
- Protactinium series $4n + 2$ (artificial branch of the uranium series)
- - - Actinium series $4n + 3$

- Bridge from uranium to actinium series
- Bridge from actinium to uranium series
- Bridge from uranium to plutonium series (Fig. 7)
- Bridge from plutonium (Fig. 7) to uranium series.

13. THE POSITION OF THE TRANSURANIUMS IN THE PERIODIC SYSTEM

Although information regarding the chemical properties of the individual trans-uraniums must still be regarded as relatively scanty, there is one fact which has now become very evident, viz. that as the atomic number increases so also does the preference of these elements for the tetravalent and trivalent states as against the mainly hexavalent uranium; and, as stated on p. 130, this preference is so pronounced when we come to element 96 that for all practical purposes we find that this element must be regarded as wholly trivalent.

Even before they had been produced in weighable quantities, it had been definitely established that these elements do not behave chemically in the same way as the higher homologues of rhenium and the platinum metals, i.e. like eka-rhenium, eka-osmium, eka-iridium and so on. Not one of them was precipitated from acid solution by hydrogen sulphide; and STRASSMANN and myself, using the 2.3-day isotope obtained by means of a high-voltage particle accelerator had very soon reached agreement with the American scientists¹ in establishing a definite similarity of element 93 to uranium. In the oxidized state, when uranium was characteristically precipitated as uranyl-sodium-acetate, it was evidently precipitated with the latter in solid solution, a fact which rendered it

¹ E. McMILLAN and T. H. ABELSON, *Physic Rev.*, 57 (1940) 1185.

easily separable from the numerous products of fission. On the other hand, it could be more easily reduced than uranium by sulphurous acid—a fact which would be utilized for separating it from uranium (see p. 109).

Long before the discovery of the trans-uraniums NIELS BOHR and other research workers had expressed the view that in the periodic arrangement of the elements, close to uranium, there might be a group of elements corresponding to the “rare-earth” group; i.e. a group based on the gradual completion of an inner electron shell and consisting of chemically similar elements. This view was confirmed by the very first experiments made with unweighable traces of element 93. It was not clear, however, where the group had its beginning. At first it was considered probable, by McMILLAN and ABELSON, the discoverers of the first neptunium-isotope, and also by STRASSMANN and myself, that the series began with uranium and that with reference to the transuraniums one might speak of the beginning of an “uranide-group”. VICTOR M. GOLDSCHMIDT and, independently, EPHRAIM considered that the series started with thorium because spectrum analysis revealed analogies between the tetravalent uranium and the tetravalent thorium. For our part we considered this improbable for chemical reasons, seeing that element 91, protactinium, which stands between the tetravalent thorium and the generally hexavalent uranium, obviously behaves as pentavalent in most of its reactions and can be separated from elements 90 and 92

much more easily than can the rare earths from one another.

After this came the discovery of elements 94, 95, 96, and recently 97 and 98, and the production of neptunium and plutonium, i.e. of elements 93 and 94, in quantities easy to deal with; the various degrees of valence and the other properties of the transuraniums were thereupon investigated with a thoroughness which would formerly not have been considered possible. Taking the views of SEABORG, it now appears most probable that the series of similar elements has its beginning as far back as element 89, the trivalent actinium.

Just as in the "lanthanides" we have the filling of the 4f-shell, in the same way, with the new elements, we obviously have the filling of the 5f-shell. The main argument in favour of the view that the series begins with the trivalent actinium is the definite tendency on the part of the higher elements to show a preference for the trivalent state.

By analogy with the lanthanide-series, this new series would have to be called the "actinide-series". The first 5f-electron would then be incorporated in thorium, the next element in the series; although here, of course, even to a greater extent than in cerium (the first member of the lanthanides) the tetravalent oxidation-state is predominant.

The deviation from trivalence, shown by the elements protactinium, uranium, neptunium and to a minor degree by plutonium, would be explained as being due to the easy separability of up to three more electrons.

It was with some reserve that SEABORG gave these details

regarding the grouping of the transuraniums into an "actinide-series". Nevertheless they appeared to him to be sufficiently well established to justify the choice of the names americium and curium for elements 95 and 96. For if we assume that the formation of the 5f-shell began at thorium, element 95 will have six 5f-electrons. The corresponding member of the lanthanide-group, europium, has six 4f-electrons. In Europe DEMARÇAY discovered europium, while in America they discovered a corresponding element 95. This was SEABORG's reason for proposing to name the latter element *americium*.

Element 96 contains seven 5f-electrons and is analogous to gadolinium in the lanthanide group. Gadolinium was so named after GADOLIN, the well-known research-worker on rare-earths. The corresponding element in the actinide-group was, by analogy, also given the name of a person, the name *curium* being chosen in honour of the two CURIES, PIERRE and MARIE, who discovered radium.

The step from the unweighable to the weighable has not yet been achieved in the cases of curium, berkelium and californium. For, as we have seen, the chain reaction which alone can produce artificial elements in quantity is maintained by the neutrons produced during fission, and it is these neutrons which, on emerging from the pile, are available in great intensity for neutron capture processes. For α -particles bombardment one still has to fall back on the cyclotron, which, while supplying α -particle beams much more energetic and more intense than those from natural α -emitters, is yet a

relatively weak source when compared with the intense neutron beam emerging from the uranium pile.

When saying this, however, we do not mean that no further isotopes of the higher elements will ever be discovered. Some neutron-reaction in the future may lead to the formation of long-lived isotopes, in which case these elements would in time also become accessible to the chemist and could be handled without danger from direct radiation, because the long-life we are assuming would involve feeble radiation.

For the time being, however, we must be content with the fact that all the gaps in the Periodic System have now been filled and that the transuranic elements of the "actinide-group" have extended the system by no less than six new elements.

14. EPILOGUE

For five hundred years the alchemists worked hard to bring about the transmutation of the chemical elements.

The first successful attempt to transform one element into another took place about thirty years ago, when research methods were transferred from the outer electron-shell to the inner atomic nucleus. Proof of the transformation was possible only by the special methods of nuclear physics, which enable one to detect the action of individual atoms. It was hopeless to think of obtaining the transformed atoms in weighable quantity. Then the giant cyclotrones were built in the U.S.A. and during the last war, for the very first time,

atoms were artificially produced in weighable quantities, even if these quantities were thousandths of a milligram or, in the most favourable case, a whole milligram.

Then the secondary neutrons produced in the fission of uranium were utilized to bring about a controlled "chain reaction" with the result that to-day representatives of nearly all the chemical elements can be produced by the gram and, in certain cases, even by the kilogram. The gaps in the Periodic System were filled; the series of elements found a continuation in the transuraniums. The long road from the alchemist's dream to the successful transmutation of many elements in unweighable quantities has now been traversed. At present, we are on the road, evidently a much shorter one, which leads to the production of artificial elements in—one is almost inclined to say—any desired quantity.

IV. SOME PERSONAL REMINISCENCES FROM THE HISTORY OF NATURAL RADIOACTIVITY

After graduating as an organic chemist and serving for two years as assistant at Marburg on Lahn under THEODOR ZINCKE, I was given the prospect of what I regarded as an attractive appointment in the chemical industry, provided that in addition to my chemistry I could offer a better knowledge of English and possibly of French, so that they could send me abroad now and then. I therefore decided to go to England for a time and left for London armed with a short letter of recommendation from my Marburg principal to Sir WILLIAM RAMSAY. While improving my knowledge of English I also hoped to do some work in chemistry.

Sir WILLIAM asked me if I would like to work on radium. When I said that I knew nothing about radium, he replied that it was an advantage to be able to approach the subject with a mind free from preconceptions. He handed me a cup containing 50 or 100 g of a salt of barium and said that the salt contained about 9 mg of radium which I was to separate from the barium by fractional crystallization and to use for preparing a number of salts of radium with the object of checking the atomic weight of the latter element. RAMSAY, famed for his work on the rare gases, had only recently become

acquainted with radium. In conjunction with F. SODDY he had proved by experiment that radium is "transmuted" into helium, RUTHERFORD having previously inferred, from his work on the electric and magnetic deflection of α -rays, that the α -rays must be helium. It was RUTHERFORD who had sent his co-worker SODDY to RAMSAY in order to test and possibly confirm his inference. The famous work by RAMSAY and SODDY was therefore the fruit of a suggestion by RUTHERFORD.

I followed the method indicated by Mme CURIE for fractionating the barium salts which had been handed to me. Very soon, however, I encountered inconsistencies which could not be disregarded. The lyes at the end of the process should have had practically no radio-activity. To my surprise, however, I found that their activity was still quite considerable and that the carrier of this activity developed not the long-lived radium-emanation of just on 4 days' half-life but the short-lived thorium-emanation of just about 1 minute half-life.

The final result of my work was therefore not the preparation, in a pure state, of presumably 9 mg of radium, but the discovery of a new "radio-element" which gave off the emanation of thorium, but was more strongly radio-active than the thorium we knew. I named the substance *radio-thorium*.

How did this substance come to be in the sample of radium? The explanation was that the sample was not extracted from a pure uranium-ore, but from an ore called thorianite, occurring in Ceylon and containing in addition to uranium a high percentage of thorium.

Strictly speaking, therefore, the discovery of "radiothorium" was just a matter of luck.

RAMSAY himself stood rather aloof from these purely radio-active investigations; the problems on which he was engaged were, of course, of a totally different nature. Nevertheless, he gave what help he could. He entrusted me, for example, with what was called a "master key" of University College so that I could enter the premises of the Chemical and of the Physical Institutes at night and carry out my experimental determinations without being disturbed, a privilege of which, for a time, I regularly availed myself.

There was another investigation which I also carried out at the RAMSAY Laboratory. This time I worked in conjunction with the late OTTO SACKUR, who unfortunately lost his life in the HABER Institute at the beginning of the first world-war in 1914. DEBIERNE, who was working with the CURIES, had found, among the hydroxides used in uranium pitch-blende processing, a new element to which he had given the name of *actinium*. This element was characterized by an extremely short-lived emanation with a half-life of only a few seconds. At the same time, the Brunswick Quinine-Factory was also engaged in preparing radium from pitchblende and it so happened that, independently of DEBIERNE, GIESEL had discovered an element which gave off a very short-lived emanation and to which he had given the name of *emanium*.

The chemical properties of DEBIERNE's actinium and of GIESEL's emanium, as originally described by the two dis-

coverers, were different, so that two different elements seemed to be involved. RAMSAY secured samples of the two substances and gave SACKUR and me a little of each. We carried out an accurate determination of the half-lives and found them both to be 3.9 sec, thus definitely establishing that actinium and emanium were one and the same element. The name actinium was retained owing to the earlier publication of DEBIERNE's investigation. It may be said, however, that GIESEL had a better knowledge than DEBIERNE of the properties—closely resembling those of lanthanum—of the new substance.

At the end of my stay in London in the summer of 1905, I was advised by RAMSAY to give up the idea of going into industry and to continue with radium research. He followed this up by writing a letter to his friend EMIL FISCHER of Berlin who thereupon offered me a place at the Berlin Chemical Institute with the prospect of a lectureship later on. First of all, however, I made up my mind to get a more thorough grounding in this new field of study by spending another winter abroad working under RUTHERFORD at Montreal in Canada; for RUTHERFORD was already regarded as the leading authority on radium. Travelling in those days was a much simpler matter than it is to-day. You simply went along to a travel-agency, picked your berth on one of the numerous German or foreign steamers and took your departure. No necessity for visas and such things!

As a kind of reference I had informed RUTHERFORD that I had discovered a new radio-active element. radio-thorium,

while working at the RAMSAY Laboratory. My application was supported by RAMSAY and a reply came inviting me to go to Montreal.

One fine day, in the autumn of 1905, I arrived in Montreal and called upon Professor RUTHERFORD. At that very first interview he made me give him an account of my "radio-thorium". He was clearly somewhat sceptical. It did not take me long, however, to convince him that radio-thorium did really exist. A few weeks later he admitted to me that in the beginning he had not believed that I had got hold of anything new. He was evidently rather sceptical regarding work done on *radioactivity* at the RAMSAY Laboratory. His initial scepticism regarding my work had been increased by a letter he had received from his friend, the outstanding radio-chemist Professor BOLTWOOD of Yale, who later on discovered ionium. It was only in after years that I learnt of this letter, written by BOLTWOOD shortly after my first publication regarding radio-thorium. Among other things the professor had written:

"The substance appears to be a new compound of thorium-X and stupidity".

I may mention that Professor BOLTWOOD and I got to know each other personally later on and that we became not only loyal colleagues, but good friends.

After a time BOLTWOOD also came round to radio-thorium. The half-life of this substance, however, became the subject of a controversy which is worthy of mention because it led,

about a year later, to the discovery of another "radio-element", namely mesothorium. From my direct measurements of radioactivity I concluded that radio-thorium must have a half-life of about 2 years. BOLTWOOD, for his part, tested a number of thorium salts and found their activity too weak as compared with the activity of the thorium minerals; it was evident that the BOLTWOOD salts contained only about 50% of radio-thorium, the substance to which the radiation of thorium is essentially due. Assuming a half-life of 2 years for the missing substance, subsequent formation should be detectable by means of an electroscope in a matter of months. BOLTWOOD, however, did not find this increase; he therefore concluded that my half-life determination could not be correct and that radio-thorium must be much more stable. As neither of us would give way it was suggested by RUTHERFORD that we should come together and argue it out. This we did and during the discussion I said to BOLTWOOD that we might perhaps reach agreement by advancing the hypothesis that between thorium and radio-thorium there occurs a longer-lived radio-element at present unknown. If in the preparation of the thorium salts the long-lived, unknown substance is separated out, this would account for the too slow or even undetectable increase in the activity of his salts. The unknown substance in question was discovered by me later on in Berlin and given the name of *mesothorium*.

I continued my work in Montreal by making a more thorough investigation of the ranges of the α -rays of radio-

thorium and of its transformation-products. RUTHERFORD cooperated with me in determining the magnetic deflection of the α -rays of actinium (and of its transformation products) and I myself, when submitting actinium to a careful chemical investigation, discovered a substance which had up to that time escaped detection, viz. *radio-actinium* (half-life about 19 days), a transition product intermediate between actinium and actinium-X, a substance which had been discovered at the RUTHERFORD Institute and in Germany by GIESEL.

I may mention that RUTHERFORD, who was no chemist; also had his doubts about radio-actinium; it did not take long, however, to bring about his conversion.

The atmosphere in the RUTHERFORD Institute was most exhilarating. RUTHERFORD had not yet become so world-famous as to attract too many pupils. Those who were with him at that time have all contributed largely towards the rapid development of research in radioactivity. There was McCLUNG, who worked on β -radiation; H. BRONSON, who carried out precision determinations of the half-lives of the transformation products of radium; A. S. EVE—RUTHERFORD's successor at Montreal—who was investigating γ -rays. RUTHERFORD himself was chiefly occupied in searching for a definite clarification of the nature of α -rays, of their deflection in magnetic and electric fields, of their absorption in their metal-foils. The only other foreigner there besides myself was Dr MAX LEVIN from Göttingen and he also was studying radioactivity; later on he became a lecturer at Göttingen but

afterwards, for extraneous reasons, had to take over his father's cloth-factory.

The apparatus we used in those days would appear very primitive to-day. We made our β - and γ -ray electroscope out of a largish tin-can or other sheet-metal box, with a smaller tobacco or cigarette-box placed on top. The leaf-holder was insulated with sulphur because in those days we had no amber.

In his α -ray experiments RUTHERFORD used a rather ancient Töpler-pump for extracting the air. The result was that in many cases the deposit under examination underwent considerable disintegration before a good enough vacuum was obtained.

On the other hand, the field of study was so new that it was an easy matter, even with primitive means, to taste the joys of discovery.

In Montreal everybody, without a trace of jealousy, recognized RUTHERFORD as the leader in scientific research. At one of the lectures held jointly with the chemists, for example, it might happen that at the end of a lecture on a subject in organic chemistry RUTHERFORD would pass a remark and then, forgetting the matter under discussion, suddenly proceed with his usual enthusiasm to deliver a lecture on his latest experiments with his beloved α -rays. Everything else was then forgotten.

RUTHERFORD's enthusiasm and abounding energy infected us all and it was the rule, rather than the exception, to work on at the Institute after supper—at least for us foreigners who

could not go on staying in Montreal as long as we wanted to. Many an evening, too, was passed at his house and most of the time, of course, we talked shop—sometimes a mixed joy for the hospitable Mrs RUTHERFORD!

It was warmly appreciated by RUTHERFORD that through Professor GIESEL he was able to purchase from the Buchler Quinine Factory in Brunswick the radium salts he required; in the beginning, indeed, at an amazingly low price. He often said that without GIESEL he would at the time never have been able to get hold of any radium. The price charged by GIESEL in the beginning for radium bromide was about 10–12 marks per mg; a few years later the price had gone up to nearly 150 marks. Later on, when he was in England, RUTHERFORD had half a gram of radium placed gratis at his disposal by the Vienna Academy of Sciences. This precious loan came to him through the good offices of STEFAN MEYER, Director of the Vienna Radium Institute.

The reader will remember the controversy over the names actinium-emanium. When I arrived in Montreal there was a controversy of the same kind going on, this time over the names *polonium* and *radio-tellurium*. Mme CURIE had described the polonium as an element chemically closely related to bismuth. MARCKWALD in Germany was also investigating the active constituents of uranium minerals and had discovered a substance with properties very like those of tellurium. He had named the substance radio-tellurium. In the end it was found that the two substances were identical. The name

radio-tellurium had therefore to be dropped, although polonium is in fact a higher homologue of tellurium. MARCKWALD did not immediately give way but after a time he also agreed that the name of radio-tellurium should be dropped. Writing to the *Physikalische Zeitschrift* on the matter, he closed his letter with the well-known quotation from Romeo and Juliet:

“What’s in a name? That which we call a rose
By any other name would smell as sweet”.

When RUTHERFORD read this he was highly delighted with what he called the “beautiful song of renunciation”. He went about in the Institute reciting the quotation in his ringing tones and long afterwards, whenever the name of MARCKWALD was mentioned, the quotation was sure to be trotted out again.

This joyous, youthful naturalness was one of the qualities that rendered intercourse with RUTHERFORD such a pleasure.

RUTHERFORD did not believe in devoting special attention to his outward appearance. As a young New Zealander he had, in the first place, gone to England with a scholarship. He had not been born with a silver spoon in his mouth. One day a representative of the English weekly *Nature* came to the Institute in order to take a photograph of the prominent authority on radium, for publication in his paper. The photo was taken in the basement room in which RUTHERFORD’s α -ray apparatus had been erected. The photographer developed the photo but regarded it as unsatisfactory. The set-up was not smart enough for the elegant English reading public;

there was not even a pair of cuffs peeping out of the jacket-sleeves. I had to lend him my own pair of "detachables". In the second photograph the amount of cuff on view still fell short of what was required. In the third, however, the cuffs came into their own and showed up in all their glory. Thus it happened that in the year 1906 I had the proud satisfaction of seeing my "detachables" immortalized in *Nature*!

In the summer of 1906 I left Montreal and, in accordance with previous arrangements, I joined the EMIL FISCHER Institute in Berlin at the beginning of the winter-semester. I had turned down the offer made by the industrial firm and had definitely been "transmuted" from an organic into a radioactive chemist.

For experiments involving measurement they placed at my disposal what had formerly been a carpenter's shop, while for the regular chemical work I was received as a guest in the private laboratory of Professor STOCK, head of the department at the Institute.

It was only natural that I should still be particularly interested in thorium and its transformation products. I got in touch with O. Knöfler and Co.'s thorium factory, which had been recommended to me by Sir WILLIAM RAMSAY, and the firm in question placed at my disposal all the chemicals and intermediate products I asked for, including, among other things, a series of practically pure thorium salts prepared at different dates. I tested their activity under conditions as identical as possible and to my delight I found a most

interesting connection between the activities and the age of the samples. Newly prepared salts exhibited an activity equal to the activity of the same amount of thorium in the mineral. As the sample grew older, its activity declined, in a few years sinking to about half the value; one of the samples, however, a salt of substantially greater age, was again showing a higher activity. What had put forward as an hypothesis in my discussion with BOLTWOOD was now translated into reality. The explanation was that the radio-thorium remained with the thorium while the latter was being prepared. With its half-life of about 2 years its activity diminished – but not indefinitely, because a hitherto unknown intermediate product of greater longevity was gradually being formed out of the thorium, the result being that in some of the older samples the activity had again gone up. The unknown substance was not difficult to discover; it proved to be in the barium.

I named the new “element” *mesothorium*. Later on I saw clearly that the radium handed to me by RAMSAY had also contained mesothorium and that the radiothorium discovered by me had first come into existence in the sample, having been formed out of the mesothorium. I could not separate the mesothorium from the radium and did not detect it.

It may be quite interesting to recall that in those days—1906 and 1907—chemists in Germany had not generally recognized and fully appreciated the important consequences of the atomic disintegration hypothesis, set up a few years before by RUTHERFORD and SODDY, and the transformation of radium

into helium, established experimentally by RAMSAY and SODDY. Now in the spring of 1907 there was held in Hamburg a Bunsen-Congress at which the main subject was radioactivity. Comprehensive treatment was given to the subject in lectures by MARCKWALD, G. MEYER, LEVIN, ENGLER and SIEVEKING, among others, and also by me. I spoke on the atomic disintegration hypothesis and its consequences. There followed a lively discussion in which, for example, Professor TAMMANN even yet declared against the elementary nature of radium and tried to advance other explanations for the transformations of radium, the formation of helium, etc. I contradicted—possibly in rather a spirited manner—because after all I had, through RUTHERFORD, attained a pretty good knowledge of the processes. In Germany, however, a young man should really have had more respect for a Geheimrat! During a break a friend of mine, MAX LEVIN, advised me to exercise a little more discretion when arguing. He had just overheard one professor ask another who I was and the other answer: “Oh, he’s one of those anglicized Berliners!”

We had never been expected to exercise discretion of that kind in democratic Canada.

EMIL FISCHER was also one of those who found it difficult to grasp the fact that it is also possible by radioactive methods of measurement to detect, and to recognize from their chemical properties, substances in quantities quite beyond the world of the weighable; as is the case, for example, with the active deposits of radium, thorium and actinium. At my inaugural

lecture in the spring of 1907, FISCHER declared that somehow he could not believe those things. For certain substances the most delicate test was afforded by the sense of smell and no more delicate test could be found than that! •

This rather reserved attitude of normal chemists towards radioactivity was maintained for quite a time. They did not take things altogether seriously. A much more progressive spirit was shown by the physicists. I can still remember the great impression made on NERNST at the Bunsen-Congress when F. VON LERCH, after mentioning the method he had devised of depositing Radium C (later on found to be an isotope of bismuth) on a nickel foil, proceeded to describe the extraordinary delicacy of radioactive measurements in the following terms: "If we divide up 1 mg of RaC among all the people living on this earth (about 2,000 million), each person would receive a quantity of the substance sufficient to discharge five electroscopes in a fraction of a second."

Further progress was made in our work at the Institute. We found that mesothorium is separable into two substances, one consisting of mesothorium 1 with a half-life of 6.7 years and not emitting any detectable rays, and the other of mesothorium 2 with a half-life of 6.2 hours and emitting β -rays; we determined more accurately the chemical properties of mesothorium and we concentrated on separating mesothorium 1 from radium. It will be remembered that the monazite used for the production of thorium contains some uranium and therewith a little radium and this fact lead the

Knöfler Thorium Factory to consider the feasibility of marketing high-activity mesothorium preparations, as substitutes for radium, which was so hard to obtain in Germany. The radium resulting as a sort of "by-product" could also be put to good use. The fresh samples of mesothorium contained, from the point of view of radiation, about 25% of radium. Owing to the formation of radio-thorium out of mesothorium 1 and 2, from now on referred to for short as mesothorium, the samples increase in strength for a year or two and then gradually decrease until after the lapse of many years only the radium is left. All my attempts to separate the two "elements" radium and mesothorium from each other resulted in failure. It proved just as impossible to separate the radio-thorium from the thorium. The chemical resemblance between the substances was clearly much greater than that, for example, between the rare earths; nobody had yet thought, however, of the possible existence of isotopes.

There was another case of surprising chemical resemblance which it will be of interest to consider rather carefully. Much attention was being given in those days to the question as to what was the direct parent substance of radium. It was known that radium occurred primarily only in uranium-ores. It was also known that the half-life of radium was about 2,000 years. Owing to the sensitivity with which radium could be detected from the emanation to which it gives rise, it could easily be calculated that the formation of radium in say 100 g of a pure radium-free uranium-salt would

certainly be detectable after the lapse of a few weeks. The experiments carried out up to this time had not produced any result. SODDY, BOLTWOOD and STRUTT had carried out experimental measurements which showed that the formation of radium out of pure uranium must certainly be more than a thousand times slower than the assumption of direct formation would lead one to expect. The inevitable conclusion from this was that between the uranium and the radium there must be a very long-lived intermediate element that separates out of the uranium during the preparation of the uranium salt (just as in the earlier case of mesothorium already described). Nobody knew what this substance was and various hypotheses such, for example, as the genesis of radium out of actinium, were put forward only to be abolished.

At the time (1907) I was still engaged on mesothorium and its properties. During the course of this work I happened to observe the presence, in a fairly large amount of a pure thorium salt, of some small amounts of radium-emanation. This was remarkable because when the pure thorium was being prepared any radium that might have been present in the thorium mineral should have been wholly extracted. A number of samples varying in age were placed at my disposal by the firm and were carefully examined by me. I found only traces of radium-emanation in 100 g of a sample which had only just been prepared; in the other samples I found that the emanation present increased with the age of the sample, the increase being directly proportional to the

age. I allowed some time to elapse and then reexamined the freshest sample and here again I found an increase. I found no increase, however, in the filtrates of the thorium precipitates. It was evident, then, that the radium parent substance for which we were looking was contained in the "pure" thorium salt, admittedly, of course, in very small quantity. The monazite sand from which the thorium had been extracted contains but very little uranium.

In order to obtain the unknown substance in greater concentration it was necessary to have actual uranium mineral to start with. Pitchblende might be the very thing. I therefore wrote to Vienna to find out if I could get a small amount of certain products which were obtainable at the Joachimsthal works run by HAITINGER and ULRICH for processing pitchblende for radium. The products I wanted were the hydrates containing the thorium portion of the mineral.

I was too late, however. Owing to the summer holidays my letter did not receive very prompt attention and in a few weeks after I had sent it there appeared in *Nature* a communication by BOLTWOOD stating that he had discovered the parent substance of radium. Its chemical properties, according to BOLTWOOD, placed it next to thorium. He named the new "element" *ionium*. My work in detecting the newly christened ionium in the pure salt of thorium, following upon work in connection with mesothorium and radium on the one hand and thorium and radiothorium on the other hand, convinced me that the chemical resemblance between ionium and

thorium must be extraordinary close. There were other cases of the same kind known to me at the time, such, for example, as the evident inseparability of radium D (radio-lead) from lead. I had not the courage, however, to conclude that in their chemical properties the elements referred to were absolutely identical.

It was left to SODDY several years later (1913) to draw from all these observations the important conclusion that there exist chemical elements with different radioactive properties and different atomic weights and nevertheless having the same chemical properties and occupying the same position in the Periodic System. To such elements SODDY gave the name of *isotopic* elements and he also applied the same conception to the ordinary chemical elements. This led to the early disappearance of the blemishes in the Periodic System and soon provided a feasible explanation of the fact that atomic weights are in many cases not whole numbers (e.g. the isotopes of chlorine). Experimental detection of "inactive" isotopes was first carried out in the gaseous element neon (THOMSON, ASTON 1913) and with the various kinds of lead of radioactive origin (HÖNIGSCHMID, SODDY 1914).

In the autumn of 1907 something happened which was to have considerable influence on my scientific development. The Viennese physicist Dr. LISE MEITNER arrived in Berlin. She had come for the purpose of extending her theoretical studies, especially by attending the lectures of Professor MAX PLANCK, and she also desired to do some experimental

work. With regard to the latter there were various possibilities open to her. As she had done some work already in Vienna involving radioactivity, she decided to keep to that branch of research and came over to me. What was originally intended to be a short visit to Berlin resulted in a cooperation and a friendship which lasted for over 30 years, in fact until the conditions prevailing in the summer of 1938 brought an end (to the cooperation, not to the friendship!)

In those days women were not allowed to work at the FISCHER Institute. When I put the matter to EMIL FISCHER, he granted permission to Miss MEITNER to work with me in the carpenter's shop on the ground-floor of the Institute, where the radioactive measurements were carried out; he requested her, however, not to enter the study rooms on the upper floor as that would be setting a precedent.

For a few years this agreement was duly observed. Later on, however, in addition to the STOCK private laboratory it became possible to fit up another part of the ground-floor for chemical research and Miss MEITNER was then able to take part in practical work of a purely chemical character such as, for example, the fractional crystallization of several hundred milligrams of mesothorium which we afterwards displayed to Emperor Wilhelm II at the inauguration of the Kaiser Wilhelm Institute for Chemistry in the autumn of the year 1912.

At the end of 1907 I was the proud possessor of an almost complete collection of radioactive "elements". At first I had

1 mg of radium; later on I received a donation of about 10 mg. In addition to the weak actinium preparations given to me by RAMSAY, I had some strongly active actinium which I had bought from Professor GIESEL. Of course I had mesothorium, also radiothorium and lastly a considerable quantities of lead chloride out of pitchblende and therefore containing radio-lead. Although it was not possible to separate the radio-lead from the enormous excess of ordinary lead, it was nevertheless possible to precipitate and to concentrate out of the lead the β -radiating RaE and RaF (polonium). Uranium and thorium were to be had in plenty, of course, and then came the "active deposits" from the parent substances radium, actinium and radio-thorium.

Having all these substances at our disposal, we decided that it would be useful to study their β -rays under identical conditions. So we put our decision into effect by undertaking in collaboration a series of investigations into the penetrating process of the rays by determining as accurately as possible their absorption in aluminium. The "half-value thicknesses" which we obtained for the rays in aluminium proved to be so characteristic for most of the decay-products that we were able to apply them in quite a number of cases as reliable touchstones in rapid analysis. We considered that we had established that under our experimental conditions the absorption of the β -rays from the individual elements proceeded in strict accordance with an exponential law and that this was a sign of their homogeneity; a crooked log. absorption

curve on the other hand, would indicate the presence of more than one element. On the basis of this hypothesis we did, in fact, find that a number of elements which had previously been regarded as non-radiating were really β -radiating elements.

In order to obtain confirmation of our views regarding the homogeneity of the β -rays, we began in 1909, in conjunction with OTTO VON BAEYER, an investigation of the magnetic deflection of the β -rays, just as RUTHERFORD had done for the α -particles of the radio-elements. The substance to be investigated was deposited on a fine wire so thinly as possible (in the case of active deposits the layer of substance was of infinitesimal thickness) and in a magnetic field those β -rays which had passed through a narrow slit were recorded on a photographic plate. The first sample investigated was the active deposit of thorium and on the plate just as we had expected, we found two and only two widely separated lines corresponding respectively to the penetrating rays of ThB (now ThC) and the softer rays of ThA (now ThB). Our assumption regarding the homogeneity of the β -rays of the individual elements had apparently been confirmed.

Our jubilation did not last long, however, On testing in this way mesothorium 2, which we had taken to be a single element, we got quite a number of fine lines. Other substances submitted to the test provided similar results and later on, when we were able to get even stronger samples of the deposit of thorium we found that this substance also gave additional lines.

Our β -ray hypothesis was therefore false. Like many another false hypothesis, however, ours gave a new trend to research—a trend which, in the course of time, found its fruition, especially later on in the work of LISE MEITNER at Dahlem and of C. D. ELLIS in England. A definite conception was formed of the difference between the band-spectra of primary β -rays and the fine line-spectra of secondary electrons released by the γ -rays from the electron-shell; and later on LISE MEITNER was the first to establish the part played by γ -rays in the creation of line-spectra.

Other experimental work went on side by side with these investigations of the β -rays of all the important radio-elements. Progress was made in the more accurate determination of the chemical properties of the products of thorium and actinium. Of course in those days nothing was known of the “isotope” conception and the tremendous simplification it introduces into the problem of arranging the numerous radioactive elements in their proper places in the Periodic System!

I will not now go into the details of this experimental work. There was one investigation, however, which deserves a special report because it provides a good example of how, in the case of radioactive measurements, minute deviations from what was expected—deviations which are regarded as of no interest owing to their minuteness—may, when submitted to careful scrutiny, lead to discoveries of considerable importance. In the years 1907 and 1908 several workers in

the field of the "active deposit" of actinium had observed an extremely attenuated residual activity the origin and nature of which they were unable to elucidate. I carried out a systematic investigation of this activity and succeeded in establishing the fact that it was due to the barium-like actinium X. At first we could not explain how this non-volatile alkali-metal could possibly get into the gaseous actinium-emanation, seeing that AcX precedes emanation in the decay-series. Raising the temperature did not cause any increase in the minute, hardly detectable amount, but we did find that the yield from thin layers of the sample was greater than that from thick layers. Just as the active deposit accumulated on a negatively charged plate so the yield from the residual activity was also greater from a negatively charged plate than from an unelectrified plate and much greater than from a positively charged plate.

Finally we took layers, about equally thin, of radio-actinium, the parent substance of actinium X, and actinium X itself, and we compared one with the other. We found clear evidence of residual activity in the radio-actinium but none whatever in the actinium X. Evidently, therefore, it was the radio-actinium that caused the residual activity; but we could not find any hitherto unknown emanation between radio-actinium and actinium. I then discovered that the explanation lay in the α -rays of radio-actinium which, like all the α -rays of radioactive substances, are shot out of the active atom with considerable kinetic energy. The explanation was this: At

the moment of its creation from the radio-actinium, the positively charged actinium X atom experiences a recoil due to the simultaneous emission of an α -particle. This recoil causes it to escape from the atomic structure, after which it travels to the negative electrode. In the case of radio-actinium the action does not in any way depend on the AcX content; the radio-actinium may be quite free from AcX. And so radioactive recoil, an effect which had been predicted years before by RUTHERFORD, was discovered experimentally. Miss MEITNER immediately suggested that we should in the same way test other α -radiating transmutation products, especially the radioactive deposits, as these could easily be prepared in layers of infinitesimal thickness. It took us only a few days to detect short-lived transmutation-products in substantial amounts, including the hitherto unknown ThD, now called thorium C'', and also actinium C'' which we had shortly before discovered in another way.

The recoil method proved of great value in the preparation of various samples in a pure state and nowadays, above all in the case of artificially radioactive substances, plays an important role especially in the preparation of radioactive indicators (SZILARD-CHALMERS method).

We also went on with our chemical researches. Our investigation of the rays of mesothorium and of radiothorium had shown that both the β - and the γ -rays of these elements have penetrative powers equal to or even exceeding those of radium and, as we have already mentioned, the thorium-

factory of Knöfler & Co. had consequently started the manufacture of strong mesothorium preparations; checking and testing were regularly carried out in our "carpenter's shop". The final chemical operations were carried out by Miss MEITNER and me in the annex to the "carpenter's shop".

These chemical separations carried out without a hood and without precautionary measures evidently caused us no harm, except for some slight damage to the fingers most exposed to the action of the rays. It must be borne in mind that the technical preparations contained about 25% of radium which came from the uranium-content of the monazite sand, so that during our work we were also continuously inhaling traces of radium-emanation.

In the year 1911 our mesothorium amounted to several hundreds mg and its strength was equal to that of pure salt of radium. Just about that period there was a great demand for radium and in addition for mesothorium, which was being supplied by the firm to hospitals etc. at about half the price of radium. The firm of Auer had also taken up large-scale production of mesothorium and up to the outbreak of the first World-War had extracted considerable quantities of this "German radium".

Another product much in demand was radio-thorium. Its decay-product thorium X and the active deposit—not so short-lived as that of radium and very easy to produce—were very suitable for application to the indicator method which had just been introduced by HEVESY and PANETH. What is

more, the α -rays of thorium C are harder than those of any other natural radio-element.

I still remember one or two amusing confusions caused by the name "mesothorium". The young ladies employed by the firm of Knöfler preferred to call it "little sunshine" because the salts, especially when strong and dry, shone beautifully in the dark (thermo-luminescence added to the normal phosphorescence). One day I was requested by EMIL FISCHER to give some information about the mesothorium rays to a Berlin Charité-Hospital professor who wanted to use them for irradiating tubercle cultures. The elderly Geheimrat received me very graciously and then began to describe the experiments he had already carried out with "semithorium". When I replied with all due modesty that the correct name was mesothorium he gave me a friendly tap on the shoulder and said: "Believe me, my young colleague, the substance is called semithorium!" Bethinking me of the bad impression I had made at the Bunsen Congress I was careful this time to correct his error with becoming modesty.

During the first World-War my colonel once introduced me to a higher officer with the words: "In civilian life Lieutenant HAHN is a professor and discovered mesothorium". The officer replied: "I believe Lieutenant HAHN is a chemist; what can he have to do with antediluvian animals?"

There were also one or two amusing misunderstandings. At a certain congress Miss MEITNER was greeted by a colleague with the words: "We have already made each other's

acquaintance". Miss MEITNER could not remember anything about it and replied: "I think you must be taking me for Professor HAHN!" Miss MEITNER evidently thought such a mistake possible in view of the fact that we had been joint authors of so many papers.

After moving out of our radioactively contaminated quarters at the Chemical Institute into the recently erected Kaiser Wilhelm Institute for Chemistry (1913) it became possible for us to do experimental work with the weakly active elements potassium and rubidium. Many years later these investigations led to what was called the "strontium method" of determining geological age—a method which was elaborated when the precision mass-spectrograph of Professor MATTAUCH was at the disposal of the Kaiser Wilhelm Institute. In the case of geologically old minerals, the strontium-method certainly has advantages over the hitherto usual lead and helium methods.

Shortly before the first World-War the most important substances in the three great radioactive decay-series, the uranium-radium, the actinium and the thorium series, had substantially been discovered and fitted into the decay-system. There was, however, one important substance still missing, —the parent substance of the actinium series.

It had long been known that actinium must be genetically related to uranium, but its direct formation from uranium had never been established in spite of its comparatively short half-life of about 15 years. This case was therefore

very like that of radium before the discovery of ionium, or of radiothorium before the discovery of mesothorium.

The problem was, in fact, an even simpler one seeing that what are called the radioactive "displacement laws" had been known since 1913; from the RUTHERFORD-BOHR model of the atom and from the works of MOSELEY it was known that there could not be more than 92 different elements. Atomic weights had given way to nuclear charge numbers or atomic numbers in the characterization of the elements. The "displacement rules" formulated by SODDY, FLECK and FAJANS enabled one to deduce directly the chemical nature of the parent substance of actinium: Actinium with nuclear charge 89 could arise either out of an element 91 by emission of α -particles (nuclear charge 2) or out of a radium isotope with nuclear charge 88 by emission of β -rays which would increase the positive charge by one unit. One short-lived representation of element 91 with a half-life of a few minutes was uranium X_2 to which its discoverer, FAJANS had given the name *brevium*. But even the discoverer of this homologue of tantalum, eka-tantalum, had failed to discover the unknown substance although he had sought for it in the right place. Shortly before the first World-War Miss MEITNER and I had also tried to find the substance by the deposition of tantalum precipitate from solutions of pitchblende. We then tried to enrich the resulting weakly α -active preparations but our method did not at first lead to any definite results. Our work was interrupted for a few years by the World-War, but in

1917 we met with some success. The substance did, in fact, prove to be a homologue of tantalum and a representative of element 91; we named it *protactinium*. We were easily able to trace the continual formation of actinium. As a spectrally pure element, it was first successfully prepared by A. VON GROSSE at our Institute. Later on several hundred mg of the pure α -radiating element were produced at the Kaiser Wilhelm Institute. Its half-life being 32,000 years, it behaves practically like a stable element; it is present in uranium-ores to about the same extent by weight as radium, but owing to its great longevity its activity is only about a twentieth of that of radium (also an α -radiating element).

I will now make brief mention of yet another substance—a substance whose importance lies in the fact that it is the first known example of “isomeric” isotopes, i.e. of those substances which are first produced in an “excited” state and then, with a definite half-life, pass over into the ground state of the atom. STEFAN MEYER had already indicated the possibility of the existence of such “isotopes of higher order”.

In this particular case I had designated the substance in question *uranium Z*; it had a half-life of 6.7 hours and was a protactinium isotope produced with a very low yield from uranium X_1 ; it is isomeric with the normal uranium X_2 which also arises directly out of uranium X_1 . What started our search for the unknown substance was the detection of a very minute discrepancy in the first results obtained for a small quantity of uranium X contained in a protactinium fraction. The

activity of the separated product amounted to only a few thousandths of the proper value for uranium X. By starting out with larger quantities of uranium we finally succeeded in extracting uranium Z in good intensity.

At first it was thought that uranium Z must be one link in a hitherto unknown uranium decay-series which could easily have been overlooked owing to its very low intensity. We sought in vain, however, for a parent substance differing from ordinary UX and isotopic with uranium X, so the only explanation left was that uranium Z is the product of a weak secondary reaction of uranium X. Many years later quite a number of such "isomers" were found among the artificially radioactive elements; uranium Z was the first one.

These researches were carried out in the early twenties. In 1919 RUTHERFORD had discovered the first transmutation of one element into another—nitrogen had been transmuted into oxygen by bombardment with α -particles.

And so a new "Age of nuclear research" had been heralded in! The age of natural radioactivity had given way to the age of artificial radio-elements. The history of this development belongs to the recent past and to the present and my personal reminiscences of earlier days have thus reached their logical conclusion.

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Chapter I is a translation of the Nobel lecture given by the author on the 13th December 1946 in Stockholm and entitled "*Von den natürlichen Umwandlungen des Urans zu seiner künstlichen Zerspaltung*" (Les Prix Nobel en 1946, pp. 167–183). It has been published by the Kungl. Boktryckeriet P. A. Norstedt & Söner, Stockholm, 1948. Chapters I and III are a combination of two of the author's publications: "*Die Kettenreaktion des Urans und ihre Bedeutung*", a detailed lecture given at the congress of the Verein Deutscher Ingenieure at Hamburg, 18–20th September 1947, to commemorate the 300th anniversary of the birth of Denis Papin (published by the Deutscher Ingenieur-Verlag G.m.b.H., Düsseldorf, 1948), and "*Künstliche neue Elemente*", based on a lecture held on the 9th May 1947 before the Gesellschaft Deutscher Chemiker at Mülheim/Ruhr, published by the Verlag Chemie G.m.b.H., Weinheim/Bergstrasse and Berlin, 1948. Chapter IV consists of a translation of the article "*Einige persönliche Erinnerungen aus der Geschichte der natürlichen Radioaktivität*", which appeared in "*Die Naturwissenschaften*", vol. 35 (1948), No. 3, pp. 67–74 (Springer Verlag, Berlin, Göttingen, Heidelberg) on the occasion of the 50th anniversary of the discovery of the radioactive elements polonium and radium by the Curies in 1898. The sections on the hydrogen bomb and elements 97 and 98 have been added to the present English edition by the author, the surveys of radioactive decay series (Figg. 7 and 8) by the editor.

