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The origins of the conceptions of isotopes

Nobel Lecture, December 12, 1922

Introduction

The work of my students and myself, for which you have so signally hon-
oured me by the award of the Nobel Prize for Chemistry for 1921, is but a
small part of much pioneering work, in many lands and stretching over a
period of nearly twenty years, into the chemistry of radio-elements and the
existence and nature of isotopes. I think I may best fulfil my duty as Nobel
laureate if I try to disentangle the real origins of the conception of isotopes,
in so far at least as I have been connected with it. What is so very simple and
clear to us today at first had a very different and puzzling character, so that
this task of tracing the birth of an idea from its earliest indications is really
both difficult and complicated. I cannot hope to achieve historical complete-
ness or finality. Rather I shall attempt to give the results of my own efforts
to trace to its beginning the idea which today is universally admitted to be of
fundamental interest and significance.

The interpretation of radioactivity which was published in 1903 by Sir Er-
nest Rutherford and myself ascribed the phenomena to the spontaneous dis-
integration of the atoms of the radio-element, whereby a part of the original
atom was violently ejected as a radiant particle, and the remainder formed a
totally new kind of atom with a distinct chemical and physical character.
These disintegrations proceed successively a large number of times, so
that there results a series of more or less unstable new elements, between the
original parent element and the ultimate unknown final product. This was a
theory sufficiently challenging to the accepted doctrines of chemistry. The
further detailed study of the chemical character of these successive unstable
elements, produced in radioactive changes, introduced an idea which was
even more subversive of the fundamental doctrines of chemistry. That idea
was that the chemical elements are not really homogeneous, but merely
chemically homogeneous. In some cases they are mixtures of different con-
stituents which. are only identical in their chemical character. Put colloqui-
ally, their atoms have identical outsides but different insides. Chemical
analysis classifies according to the external systems of electrons which sur-

round a small massive internal nucleus, whereas radioactive changes, which are of the character of veritable transmutations, concern the internal constitution of this inner nucleus. They showed that the same exterior may conceal very different interiors in the atomic structure. These elements which are identical in their whole chemical character and are not separable by any method of chemical analysis are now called isotopes.

I may begin with a brief statement of the earlier researches into the nature of radioactivity. The power of spontaneously emitting rays of a new kind was discovered in 1896 by Henri Becquerel for the compounds of the element uranium. The physicists sorted the rays emitted into three types, the α−, β− and γ - rays, and their real nature was quickly elucidated. The β -rays were shown to be due to the expulsion of negative electrons traveling at a far higher speed than any that can be artificially produced in the vacuum tube as cathode-rays. The γ-rays were correctly regarded as X-rays, but of a greater penetrating power. In due course the X-rays were shown to be waves of light of extraordinary short wavelength and high frequency, whilst the γ-rays are also of the nature of light, but of even shorter wavelength and higher frequency. The α -rays were first proved to be due to radiant atoms of matter carrying a positive charge and were identified, after many years of continuous work by Sir Ernest Rutherford, with helium atoms carrying two atomic charges of positive electricity. Sir William Ramsay and I had shown in 1903 that helium was being continuously generated from radium in a spectroscopically detectable quantity, and subsequent work showed that helium is generated, as α-particles, in all the radioactive changes where α− rays are expelled.

The nature of the ionization produced in gases by all these new types of radiation was quickly elucidated, and as a consequence highly sensitive and accurate methods of measurement were evolved, which, more than any other single factor, contributed to the rapid development of the subject.

On the chemical side, the work of M. and Mme. Curie had shown radioactivity to be a specific atomic property, definitely restricted to the last two of the then known elements, uranium and thorium, though the elements potassium and rubidium emit β-rays, exhibiting in this respect evidence of true radioactivity. Using the new property much as the pioneers with the spectroscope used the spectra in the discovery of new elements, they discovered the powerfully radioactive new elements, polonium and radium, and, M. Debierne, actinium, in the uranium minerals. Of the many similar new radio-elements now known radium is still the only one the compounds of

which have been prepared in a pure state, and for which the spectrum and atomic weight have been determined. As regards its whole chemical character radium is absolutely normal and its properties could have been predicted accurately for an element occupying its place in the Periodic Table. It is the last member of the family of alkaline earths and stands in relation to thorium as thorium does to uranium in the periodic system. But superimposed on this normal chemical character it possesses a radioactive character truly astounding. The energy it evolves in the form of the new radiations, measured as heat, amounts to 133 calories per gram per hour. The theory of atomic disintegration shows however that the uniqueness of radium is due mainly to the fact that its average life, 2440 years, is sufficiently long to allow the element to accumulate in the minerals in which it is formed to a ponderable quantity, and yet short enough for the rate at which the energy of disintegration is liberated to be truly surprising. In the elucidation of the various disintegration series of uranium, thorium and actinium, every degree of atomic instability is encountered. The average life varies from the order of 1010 years, for the primary radio-elements uranium and thorium, down to periods of the order of a minute, which marks the limit beyond which the chemical character cannot be investigated. But by physical methods members down to a period of 1/350 second have been put in evidence, and two, of the order of 10^{-6} and 10^{-11} second, are indirectly inferred.

But for all these the theory indicates, as for radium, a perfectly normal and definite chemical character as well as the superimposed radioactive character. There is no progressive change in the nature of the atoms. As they were when produced, by the explosion of the atom from which they originate, so they remain till they in turn explode. Otherwise the law of change found to hold could not apply. The chemical and spectroscopic character is that of the atom during its normal and uneventful life, whilst its radioactive character is that produced by its sudden death. But for this very definite and precise implication from the theory of atomic distintegration it might have been supposed that the chemical character of such extraordinary substances would also be extraordinary.

Possibly one other introductory remark may be justified, as to the kind of evidence upon which the theory of atomic disintegration was originally founded. In investigating the radioactivity of thorium compounds, we found that a constituent responsible for the greater part of its radio-activity could be separated by the use of specific reagents. Thus when thorium is precipitated by ammonia, the thorium X remains in the solution, though

when other precipitating reagents, such as ammonium carbonate, oxalate or phosphate, are employed no separation is effected. But this thorium X as fast as it was removed reformed at a perfectly definite rate. Now if a chemist were to purify, say, lead from silver and found as often as he purified it that the silver reformed, he would have to conclude that lead was changing into silver. The fact that this does not happen is at the basis of the belief in the unchangeability of the elements. So the conclusion that in radioactivity the elements are changing into others is equally direct. It is not a valid criticism that the products are usually too small for chemical examination. If the chemist referred to found that his lead after purification in due course showed again all the usual analytical reactions for silver he would be perfectly satisfied, though anyone else might reasonably ask to be shown a shilling made by the process and vouched for by the Mint. Radioactive character provides as fine a criterion for quantitative and qualitative analysis as any of the chemical and spectroscopic tests depended upon by the chemist. It is as idle to expect the radio-chemist to produce his materials in quantity sufficient for chemical examination before forming conclusions, as to expect an analyst in the course of his work to separate and prepare the various constituents of the materials he analyses in form and quantity capable of satisfying a jury with no chemical training.

The experimental method that first revealed isotopes

The history of isotopes fittingly commences with the discovery of radiothorium, a new product in the thorium disintegration series, by Sir William Ramsay and Otto Hahn in 1905, which is intermediate between thorium and thorium X and generates the latter in the course of an α-ray change. A considerable quantity of thorianite, a newly discovered mineral with 60 to 70% of ThO_2 and 10 to 20% of U_3O_8, was worked up for radium by the known methods. The new substance was discovered during the fractional crystallization of the barium-radium chloride and was separated from the radium along with the inactive barium in the course of the fractionation.

In 1905 also the discovery by Godlewski of actinium X, an intermediate product between actinium and its emanation, in every way analogous to thorium X, was the commencement of the recognition of the general analogies that existed in the three series. Even so early as 1905 I said*:

*In order as far as possible to avoid the charge of reading into this account of the

"The general resemblance in character and relative period of the succeeding members of the various disintegration series on the one hand, and their distinct individual peculiarities on the other, are most fascinating and mysterious. We seem to have here an extension of similar relations expressed for elementary matter generally by the periodic law, and there is no doubt that any explanation of the one will have an application to the other." (1905, p. 308).

Reverting now to radiothorium, all attempts to separate it from thorium compounds either failed completely or seemed successful to only a minute extent. In 1906 Elster and Geitel and also G. A. Blanc, after having discovered radiothorium as a constituent responsible for part of the radioactivity of the sediments from certain hot springs, used the experience they had gained in its separation in attempts to separate radiothorium from thorium compounds, and were, apparently, to a very slight extent successful. Dadourian, Boltwood and McCoy and Ross found that commercial thorium salts contained only one-half as much radiothorium as the mineral from which they extracted and as the thorium they themselves separated from minerals in the laboratory. This seemed to indicate that the secret operations by which thorium is commercially extracted were much more effective in separating radiothorium than the known laboratory methods, but a search of the by-products of the thorium manufacture failed to locate the missing radiothorium.

In 1907 Hahn discovered mesothorium, a product intermediate between thorium and radiothorium and generating the latter. In 1908 he showed that mesothorium itself consisted of two successive products. The first, mesothorium 1, is produced directly from thorium and has an average life of 9.67 years. It is one of the two radio-elements, actinium being the other, which disintegrates without the expulsion of any detectable radiation, though in all probability a β-particle is expelled, but its velocity is too low for it to be detectable. Its product, mesothorium 2, gives powerful β–and γ-rays and has an average life of 8.9 hours.

In the meantime it was found that the radioactivity of the thorium compounds prepared in the laboratory from the mineral decayed during two

history of isotopes more in connection with each advance than the state of knowledge at the time warranted, I shall frequently quote literally my *Annual Report on Radioactivity,* which I have contributed, *since* their commencement in 1904, to the *Chemical Society's Annual Reports on the Progress of Chemistry,* with the date of the Report and page from which the quotation is taken in brackets at the end of the quotation.

years from preparation to one-half and became the same as that of the commercial preparations, whereas the latter increased in activity with age and the older they were the more nearly their activity became equal to that when freshly prepared from the mineral. In the process of separating the thorium, mesothorium, but not radiothorium, is separated. The latter, pending the regeneration of the former, at first decays, and subsequently, as its parent, mesothorium, begins to reaccumulate, is regenerated until finally it reaches its original equilibrium value.

"The effect then had nothing to do with the method of preparation of the thorium salt, but is purely a question of age." (1907, p. 325).

It was the first effect to be recognized of many similar ones subsequently. For example in 1911 Antonoff discovered uranium Y, and atributed his success in its separation to the particular chemical processes adopted, whereas it was in reality due to the lapse of a suitable period of time between successive separations.

Boltwood concluded from his experiments that mesothorium must be removed from thorium along with thorium X when thorium is precipitated by ammonia, the original method by which thorium X was discovered.
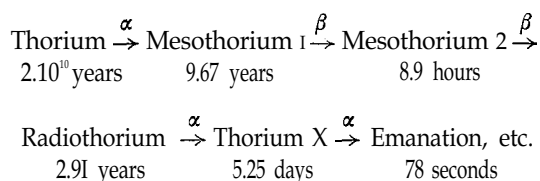
This year, 1907, marks the first definite statement of the doctrine of the complete chemical non-separability of what are now called isotopes. McCoy and Ross[1] after describing many careful and prolonged attempts, all of them completely without success, to separate radiothorium from thorium, said:

"Our experiments strongly indicate that radiothorium is entirely inseparable from thorium by chemical processes"; and they drew from this the correct inference with regard to the radiothorium separated by Ramsay and Hahn and others: "The isolation of radiothorium from thorianite and from pure thorium nitrate... may have been accomplished by the separation of mesothorium, which in time changed spontaneously into radiothorium."

Thus already in 1907 the experimental method that first revealed the existence of isotopes among the successive products of radioactive change had been applied and its implications well understood.

"Although the separation of mesothorium from thorium and of thorium X from radiothorium is easily accomplished, there is no known method of separating by chemical means either radiothorium from thorium or thorium X from mesothorium. But, owing to these last mentioned pairs alternating in the disintegration series, each of them can readily be prepared by itself. The preparations of radiothorium obtained by Hahn and others are probably not the radiothorium existing in the original mineral, but regenerated radio-

thorium subsequently produced, after the separation, from the easily separated mesothorium." (1907, p. 326).

The first part of the thorium disintegration series is shown below with the average life of each member and the rays expelled in its change.

$$\text{Thorium} \xrightarrow{\alpha} \text{Mesothorium I} \xrightarrow{\beta} \text{Mesothorium 2} \xrightarrow{\beta}$$
$$2.10^{10}\ \text{years} \qquad 9.67\ \text{years} \qquad\qquad 8.9\ \text{hours}$$

$$\text{Radiothorium} \xrightarrow{\alpha} \text{Thorium X} \xrightarrow{\alpha} \text{Emanation, etc.}$$
$$2.9I\ \text{years} \qquad 5.25\ \text{days} \qquad 78\ \text{seconds}$$

The separation of thorium and radiothorium and of mesothorium I and thorium X by chemical analysis, now as then, are completely impossible. But each can be obtained alone by a suitable combination of chemical processes at suitable intervals of time. The exact procedure is dictated by the relative periods of the substances to be separated. In this connection it must be remembered, since the methods of ascertaining the nature of the products separated are purely radioactive, that it is the relative intensity of the radioactivity, or the relative number of α– or β–particles emitted per second, and not the relative weights, which is of importance. For different radioactive elements, the radioactivity is proportional to the weight divided by the period. A rapidly changing substance reforms and attains its equilibrium value correspondingly quickly, a slowly changing one correspondingly slowly.*

The ease with which the mesothorium is separated, and the very long time required for it to reform in substantial amount, is the reason why it was not discovered in 1902 when thorium X was discovered. It would normally
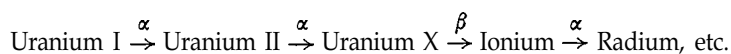
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* Thorium may be obtained, free from radiothorium, by repeated precipitations with ammonia over an interval sufficient to allow the original radiothorium present to decay. Mesothorium 1 is obtained free from thorium X from the first filtrate in the ammonia precipitation after leaving it for a month for the thorium X to decay. Radiothorium is obtained free from thorium, by separating the mesothorium, waiting for it partly to change into radiothorium, and then separating the radiothorium from the mesothorium, by adding a trace of aluminium or similar substance to serve as a vehicle in the filtration, and precipitating it with the radiothorium by ammonia. Lastly thorium X is obtained free from mesothorium either from radiothorium or from thorium by the ammonia precipitation, in the latter case from the second and successive precipitations at monthly intervals, the first filtrate containing the mesothorium.

be present only in thorium compounds that have remained undisturbed a long time since preparation, or purification. This fact had later an important sequel, as we shall see.

The question, whether these non-separable pairs of radio-elements are really chemically identical or not, is not of importance in the argument. What is important is that 15 years ago pairs of radio-elements, actually not separable by chemical analysis, were in fact separated by successive chemical analyses at suitable intervals. If these pairs had been consecutive in the series, instead of being separated by intermediate products of different chemical character, they must have remained unresolved. Such a case, it seemed, was known.

The isotopes of uranium

In 1908 Boltwood showed that the relative α–ray activities of the various α–ray giving products of the uranium series, in equilibrium in natural minerals, conformed to the expulsion of one α-particle per atom disintegrating, except in the case of the actinium series (which so was correctly indicated to be a quite minor branch of the main uranium-radium series), and *uranium itself* which gives two α-particles per atom disintegrating. This satisfied the atomic weight difference of 12 units between uranium and radium, which corresponds with the expulsion in all of 3 α–particles. Ionium, a long-lived intermediate product giving α-rays and generating radium, accounts for the third α-particle. Thus it appeared that the 2 α– particles from uranium were due to two successive α–ray changes

$$\text{Uranium I} \xrightarrow{\alpha} \text{Uranium II} \xrightarrow{\alpha} \text{Uranium X} \xrightarrow{\beta} \text{Ionium} \xrightarrow{\alpha} \text{Radium, etc.}$$

and that uranium I and uranium II were consecutive products, completely similar in chemical character, analogous to thorium and radiothorium, except that there was no intermediate product of different chemical character to serve for their separate recognition.

The correctness of the inference that the two α-particles are successively and not simultaneously expelled was established by Marsden and Barret in 1911. They observed the scintillations produced by a uranium compound on a zinc-suiphide screen, and found no pairs, or any preponderance of short intervals between scintillations, beyond that required by the theory of

probability for a purely random distribution of intervals. Although the other inference, that the two uraniums are consecutive successive products, was theoretically incorrect, because it was later found necessary to place uranium X between the two uraniums, the practical consequences are unaffected. Owing to the relatively short period of uranium X in comparison with its product uranium II, the latter cannot be put into evidence, as radiothorium was, by separating its parent and allowing it to change. The quantity of uranium II so formed (divided by its period) is too small to be detected, even when the uranium X from 20 kilograms of the element uranium is studied. Even if sufficient uranium could be experimented with, the experiment would still be rendered difficult, if not impossible, because of the ionium in the series, which is much shorter-lived than uranium II, and non-separable from uranium X, just as thorium X is from mesothorium I. This then is a case to which the experimental method of separation does not apply owing to the hopelessly unfavourable relation between the periods of the successive products.

The chemical identity of different radio-elements and its implications

Examples of chemically non-separable pairs, or groups, of radio-elements now began to accumulate very rapidly. Ionium, discovered by Boltwood, and by Marckwald and Keetman independently, in 1908, which proved to be the direct parent of radium, was shown to be identical in its chemical character with thorium. Boltwood separated it from uranium minerals by adding a little thorium and by separating and purifying the latter. Marck-wald and Keetman obtained it by precipitating the rare earths in the mineral with hydrofluoric acid, and found that any of the regular methods for purifying thorium from the rare earths would separate the ionium from the latter and actinium, but not from thorium. Keetman[2] tried twelve good methods, all well known to be effective in the purification of thorium, without effecting the least separation of ionium. He found at the same time that he could not separate uranium X, either, from the ionium-thorium mixture, though here, of course, just as with thorium X and mesothorium, the uranium X, being short-lived, speedily disappears of itself.

Auer von Welsbach also, in 1910, carried out a masterly technical separation of the ionium and actinium of the "hydrate" fraction obtained from 30,000 kilograms of Joachimsthal pitchblende in the manufacture of radium,

and prepared a very concentrated ionium preparation, upon the spectrum and atomic weight of which other investigators subsequently worked. He failed completely to separate the ionium from the thorium, though many fresh methods were tried.

Herschfinkel tried numerous methods to concentrate radium D (radio-lead) from lead and failed completely. Three years later, Paneth and von Hevesy tried twenty different methods, also completely without result, and upon this failure they based their valuable and elegant method of using the radio-elements as indicators. For example, in the present case, the solubility of very insoluble lead compounds was determined by adding radium D *to* the lead, and determining the almost unweighable quantities of lead dissolved by radioactive methods. This use has wide applicability in chemical problems. In this instance, radium D can be obtained free from lead, if required, by sealing up the radium emanation in a capillary tube and allowing it to change. After a month, the conversion of the emanation into practically pure radium D is complete. In this way they were able in 1914 to extend the chemical identity between radium D and lead to its whole electrochemical character.

In contrast with the foregoing cases, the cases of polonium (radium F) and actinium may be mentioned. Although these radio-elements have never yet been isolated in the pure state, nor even their spectra yet determined, this is solely, probably, on account of their relatively short period, and consequently infinitesimal quantity. Thus polonium was shown by Mme. Curie to resemble bismuth closely, and by Marckwald tellurium. But it may readily be concentrated by chemical methods from both bismuth and tellurium. Actinium was shown by Giesel to resemble lanthanum most closely of the rare-earth elements, but he succeeded in effecting a partial concentration of actinium from lanthanum.

In 1909, Sweden made a very notable contribution to this subject in the work of Strömholm and Svedberg, who made use of isomorphism as a means of ascertaining the chemical character of the radio-elements. They crystallized in the solutions of the radio-elements different salts and determined whether or not the radio-element crystallized out with the salt. Thus they found that thorium X crystallizes with lead and barium salts, but not with others. They correctly characterized it as an alkaline-earth element, in contradiction of an earlier and incorrect conclusion that it was monovalent, arrived at from a study of its diffusion coefficient and ionic mobility. Actinium X was also characterized as an alkaline-earth element, and they were

unable to find, by these methods, any differences in the chemical behaviour of radium, thorium X and actinium X, even in their quantitative aspect.

Strömholm and Svedberg were probably the first to attempt to fit a part of the disintegration series into the Periodic Table. They were correct as regards thorium X and actinium X, but were misled by a faulty characterization of the chemistry of mesothorium I. It is probable that the reason for this was that already referred to, which prevented mesothorium being discovered when thorium X was, namely, that it has been unwittingly separated in the preliminary purification of the material. Nevertheless, in their conclusion, is to be found the first published statement, that the chemical non-separability found for the radio-elements may apply also to the non-radioactive elements in the Periodic Table. Remarking on the fact that, in the region of the radio-elements, there appear to be three parallel and independent series, they then say "one may suppose that the genetic series proceed down through the Periodic Table, but that always the three elements of the different genetic series, which thus together occupy one place in the Periodic System, are so alike that they always occur together in Nature and also not have been able to be appreciably separated in the laboratory. Perhaps, one can see, as an indication in this direction, the fact that the Mendeleev scheme is only an approximate rule as concerns the atomic weight, but does not possess the exactitude of a natural law; this would not be surprising if the elements of the scheme were mixtures of several homogeneous elements of similar but not completely identical atomic weight." Thus Strömholm and Svedberg were the first to suggest a general complexity of the chemical elements concealed under their chemical identity. Until I read their paper again, in the preparation for this lecture, I had not realized how explicit this anticipation of present views is.

In 1910 a new and, as it proved, very important case of chemical identity was discovered, that between mesothorium I and radium. This was published independently by Marckwald and myself, though it must have previously been known to Hahn and those engaged in the technical extraction of mesothorium, who, however, kept it secret. Marckwald made the discovery through being asked to examine a radium preparation prepared from a thorium-containing uranium mineral, which owed the greater part of its activity to mesothorium. I worked it out independently for preparations I obtained from thorianite. Contrary-to the impression, which then prevailed, that barium sulphate precipitated in the solution *entrains* or *adsorbs* mesothorium, as it does uranium X, I found it impossible to separate the

mesothorium from the radium in the fractional crystallization of chlorides of mesothorium, radium and barium, which I so obtained. The radioactive constituents were concentrated from the inactive barium, to an extent of several hundred times, without the slightest change in the ratio between them, so far as could be ascertained by the most careful radioactive measurement. Thorium X was also found to be non-separable from mesothorium and radium in agreement with Strömholm and Svedberg's conclusion. In the fractional crystallization Ramsay and Hahn found the radiothorium to be separated with the barium. Their discovery of this element was entirely due to the fact that between the separation of the radium with the mesothorium, a certain, probably considerable, interval of time must have elapsed.

From this date I was convinced that this non-separability of the radio-elements was a totally new phenomenon, quite distinct from that of the most closely related pairs, or groups, of elements, hitherto observed in chemistry, and that the relationship was not, as usually supposed, one of close similarity but of complete chemical identity.

"When it is considered what a powerful means radioactive methods of measurement afford for detecting the least change in the concentration of a pair of active substances, and the completeness and persistence of some of the attempts at separation, which have been made, the conclusion is scarcely to be resisted that we have in these examples no mere chemical analogues but chemical identities." (1910, p. 285).

The case of the rare-earth elements has often been cited as an example of mixtures of elements, once thought homogeneous, but with the application of more refined methods, or more repeated separations, proving to be separable. But the case is not really comparable. The difficulty in the rare earths is not so much in the actual separation, but in ascertaining what the effect of an attempted separation has been. Every process has to be laboriously repeated a large number of times before a separation detectable by the insensitive methods available, such as the determination of the chemical equivalent, is apparent. So soon as any difference in properties is detected, separation follows. Thus, didymium was resolved owing to the difference in colour of the salts of its two constituents, and now the spectra afford a more general cxiterion of the same character. But for the radio-elements the substances are known severally as individuals from the start, unrivalled methods are available for detecting the least beginnings of separation so soon as it occurs, and yet the constituents once mixed remain inseparable by chemical processes[3]. Today, of course, no argument as to the complete chemical iden-

tity of isotopes is required, since half the common elements examined by the positive-ray methods have been found to be mixtures of isotopes.

On broader and quite general philosophic grounds, and without in the least postulating a continuation of the genetic series of the radio-elements throughout the Periodic System, I arrived at the conclusion reached by Strömholm and Svedberg. It was certain that among the groups of chemically identical radio-elements differences of atomic weight of whole units must exist. Thus from the atomic weights of the parent elements and the number of α-particles expelled, the atomic weights of ionium (230) and radiothorium (228) must differ by two and four units, respectively, from that of the chemically identical thorium (232). Once one enquired what evidence the chemist had for the real homogeneity of the elements as distinct from their chemical homogeneity, the conclusion followed at once that if all the elements were mixtures in constant proportions of chemical identities differing step-wise by whole units in atomic mass, the chemist with his methods must have remained unaware of it.

"The recognition that elements of different atomic weight may possess identical chemical properties seems destined to have its most important application in the region of the inactive elements, where the absence of a second radioactive nature, totally unconnected with the chemical nature, makes it impossible for chemical identities to be individually detected. Chemical homogeneity is no longer a guarantee that any supposed element is not a mixture of several of different atomic weight or that any atomic weight is not merely a mean number." (1910, p. 286).

The spectrum of ionium

In 1912 Exner and Haschek, using the ionium-thorium preparation of Auer von Welsbach, already alluded to, and A. S. Russell and Rossi, using an active ionium-thorium preparation prepared for the Royal Society, independently attempted to determine the spectrum of ionium, without, in either case, succeeding in finding a single new line. The spectra were those of thorium simply.

The interpretation of this remarkable result was, at the time, dependent upon the incompletely known period of ionium. This constant fixes the proportion of ionium by weight in the materials used. All the evidence was in favour of ionium having a very long period, so that the proportion of

ionium by weight in both preparations would be so high that the failure for it to show any spectrum due to ionium would be offundamental importance.

The low range of the α-rays from ionium indicated a period for this element of the order of from 2 to 5 x 10^5 years, but this evidence is but slight. On the other hand I had been engaged continuously since 1905 in an experimental effort to detect the growth of radium in carefully purified uranium. The failure to detect this after many years led to the minimum estimate for the average life of ionium of 10^5 years. This depended on one condition, that ionium was the only intermediate of long period between uranium and radium (uranium I and II for this purpose being considered a single element). This has now been established. Over sufficiently long periods the growth of radium from uranium has been found to proceed according to the square of the time. This proves that ionium is the only long-lived intermediate. Also the actual period of ionium to an accuracy of two per cent has been found to be 10^5 years. So that a considerable percentage of the ionium-thorium preparations must have been pure ionium. Exner and Haschek concluded that the period of ionium must have been much overestimated, and that this appeared from their result to be less than that of radium. But they realized the impossibility of reconciling this with the other evidence. Russell and Rossi, after reviewing all possible explanations, remarked: "But the possibility that they (thorium and ionium) are identical in all physical and chemical properties, and differ only in atomic weight and radioactive properties, should not be lost sight of."

I endorsed this view:

"The simplest, if somewhat heterodox, view to take is that ionium is a long-period element, and that its spectrum, as well as its whole chemical behaviour, is identical with that of thorium. It is clear that the conception of the chemical elements as necessarily homogeneous is undetermined and that different elements with different atomic weights are chemically identical not as an exception but as a consequence of the way in which the known disintegration series have been shown to run their course. In non-radioactive matter this heterogeneity cannot be distinguished, but it is not so in the case of matter actually in process of evolution." (1912, p. 322).

The radio-elements and the periodic law

I now pass on to the second phase of this history, the fitting of the radio-

elements into the Periodic classification. In 1911 in the first edition of my *Chemistry of the Radio-elements* (pp. 29 and 30) I discussed the effect on the chemical character of the loss of the α-particle, which is a helium atom carrying two atomic charges of positive electricity.

"The loss of a helium atom or α-particle appears to cause the change of the radio-element not into the next family but into the next but one... Thus we have in each series a well-marked sequence from the tetravalent family (radiothorium, ionium) into the divalent family (radium, etc.) and into the non-valent family (emanations). Again the product derived from polonium is probably not bismuth but lead, in each case the step being from the family of even valency into the next, the family of odd valency being missed. But this is not all. The progress is certainly not so straightforward as this. In several cases the matter appears to *alternate* in its passage, passing through the same family not once but *twice.* The product from thorium (group IV) is mesothorium (group II). The product of the latter is radiothorium (group IV), which in turn produces thorium X (group II). Again radium D is chemically non-separable from lead (group IV), its product is polonium (group VI), while the product of the latter is almost certainly lead (group IV)."

This α-ray generalization enabled me to see that uranium X must be intermediate between the two uraniums (1912, p. 321) though his conclusion was published by several others as well as by myself before the Report was issued.

But I was at fault in one point, in that I thought the "active deposit" products, such as radium A, B and C, were unstable transition links between the beginning of the period and the end of the last, analogous to the VIIIth group between gold and tungsten, and not represented in the table.

In 1912, the chemical character of the post-emanation, or active deposit, products began to attract more attention. The ease with which these products are separated from all inactive matter from the volatile emanations, and, after the discovery of recoil methods, partly from one another, without chemical methods accounted probably for the fact that none of them were known to resemble known elements. However, von Lerch from as far back as 1905 had studied their electrochemical deposition. His rule that the successive products are successively more electrochemically "noble", i.e. more easily deposited on metals, and on the cathode during electrolysis, in acid solution, though true for radium A, and for the B and C members, was found to fail for thorium D, which is less easily deposited than thorium B.

Rutherford in his original discovery of the thorium active deposit, had shown that it volatilized at a bright red heat. Makower in 1909 had found for

three constituents of the radium active deposit, the following volatilization points, radium A 900°, radium B 600° and radium C 1200° C. In 1912 Schrader and, after him, A. S. Russell, observed that the volatility of the active deposit is much affected by exposure to various gaseous reagents such as chlorine, and by volatilizing in different atmospheres. Thus in hydrogen radium C commences to volatilize at 360° and, for the three products together, volatilization is complete at 650°. This clearly pointed to definite chemical and physico-chemical properties even of these excessively ephemeral elements, which before had been thought to exist in too infinitesimal quantities to show such definite reactions. In this year, von Hevesy studied the electrochemistry of these products from the standpoint of modern theory, and showed that they might be used to test the consequences of this theory at degrees of concentration far below anything that can be attempted by any other method. He also obtained the important result that in their electrochemical behaviour the three B-members are identical among themselves, and also the three C-members.

In 1912, A.Fleck, commenced to publish the results of a long series of systematic studies into the chemistry of such of the radio-elements, with period over one minute, as remained uncharacterized, from the definite point of view of finding which known element each most resembled in chemical character, and then whether it was separable or not from that element when mixed with it[4]. He confirmed the characterization of uranium X as chemically non-separable from thorium, showed that radioactinium, which Strömholm and Svedberg had found to be isomorphous with thorium, was in fact non-separable from it, and, most important of all, proved that thorium B is non-separable from lead. Hitherto from von Lerch's electrochemical researches it had been supposed that these products were chemically allied to the noble metals. In his next communication, Fleck showed that mesothorium 2 was non-separable from actinium, that radium B and actinium B were extremely similar to lead and probably non-separable from it, that the three C-members are closely allied and probably chemically similar to bismuth, whilst radium E has properties in all respects identical with those of bismuth[5]. These results, as concerns the B- and C-members and radium E, were at variance with those found at the same time by von Hevesy[6] from the speed of diffusion, a method that previously had given a fase result for thorium X. Hevesy classed the B-members as univalent and the C-members and radium E as divalent, but later showed these conclusions were in error.

From now on, the history of this development is extraordinarily involved, and brief reference to the actual dates of the various publications is essential before passing on to the nature of the advance itself. G. von Hevesy and A. S. Russell were both working in Rutherford's laboratory in Manchester, and published within a fortnight papers containing a form, only in part correct, of the great generalization, which was to clear up, as in a sudden flash, the whole subject. A fortnight after Russell, K. Fajans working in Karlsruhe, published this generalization complete and accurate in every detail, A fortnight after Fajans, in a paper amplifying and amending Russell's scheme, I published exactly the same scheme as that of Fajans, whose paper only reached England after mine was written. His generalization was remarkable, in that at the dates on his two papers Dec. 31, 1912 and Jan. 15, 1913 only one of Fleck's important results, that of thorium B, had been published. But to me in full knowledge of all Fleck's experimental results, the complete generalization was obvious so soon as Russell's paper directed attention to it. Till then I had scrupulously avoided even thinking about these results until they were completed, for fear of giving an unconscious bias to the work in progress in the laboratory.

Hevesy[7], from a general examination of the valencies of the radio-elements, concluded that the effect of the expulsion of the p-particle is to alter the *valency* by one in the opposite direction to the effect of the expulsion of the α-particle, which produces an alteration of two. But for the pre-emanation members, the direction in which the valency was altered was regarded as the opposite of that for the post-emanation members. He thus supposed that the valency, having been reduced to zero at the emanations by successive a-ray expulsions, then rises to two, making radium A divalent. This got the tetra-valent elements right, but others ($RaCa^2AcD$, and ThD) were wrong. Others again could be wrong or right according as the valency is taken as the Group Number or 8 minus the Group Number.

Russell* dealt with the position of the element in the Periodic Table, and not with its valency, and published the first scheme tracing the whole disintegration series through the table. In the pre-emanation part he predicted the existence of uranium X, in the vacant place between thorium and uranium, and was in general correct, showing the effect of the expulsion of the $\beta-$ ray as causing *a shift of one place in the direction opposite* to the double shift caused by the expulsion of the *a-ray.* But he failed to envisage the Periodic Table as a continuous succession of places, and passed from the emanation or zero family to the preceding members in the table in the reverse order,
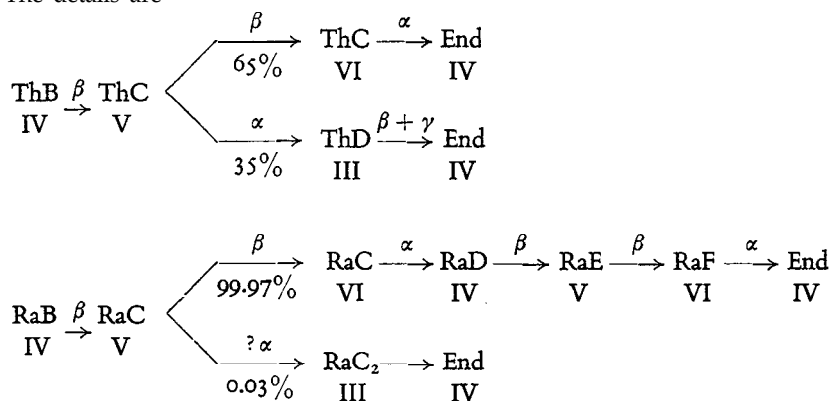
placing radium A with mercury. Though he got the C-members correct
to Fleck's characterization, it was by arbitrarily making the effect of the β−
ray in the same direction as that of the α-ray change*.

   In the correct generalization of Fajans[9] and myself[10], each a-ray expelled
causes a shift of two places in the direction of diminishing mass and diminish-
ing valency, as I had pointed out two years before, and each β-ray expelled
causes a shift of one place in the opposite direction, consistently throughout
the whole series, including the very complicated branchings that occur at the
C-members**. In the elucidation of these, Fajans had himself been the pio-
neer the year previously.

   The scheme is shown in Fig. I (from the British Association Reports,
Birmingham Meeting, 1913) in substantially the form in which I first pub-
lished it, except that the actinium series has been separated to make it clear-
er. In its present form (1922) it is shown in Fig. 2 (from a recent paper on
Isotopes, contributed to the Brussels Conference of the Institut Internatio-
nale de Chimie Solvay, April 1922), which shows the actinium series linked
up with the uranium series***. In the first (Fig. I), the places in the Periodic
Table are represented as differing by one in the number of electrons in the
atom, a loss of one electron being regarded, purely conventionally, as equiv-
alent to the gain of a single atomic charge of positive electricity. This was
shown only in the diagram in the *Jahrbuch der Radioaktivität,* not in that

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* Used as we now are to the idea of Atomic Number, this error may now seem obvi-
ous. Rather it is the great clarification of the nature of the Periodic System, first effected
by this very generalization, which makes the error appear obvious.

** The details are

$$ThB \xrightarrow{\beta} ThC \begin{cases} \xrightarrow[65\%]{\beta} ThC \xrightarrow{\alpha} End \\ \xrightarrow[35\%]{\alpha} ThD \xrightarrow{\beta+\gamma} End \end{cases}$$

ThB β ThC
 IV → V
                    β
                ───────→ ThC ──α─→ End
                 65%   VI   IV

                    α   β + γ
                ───────→ ThD ──────→ End
                 35%   III   IV

RaB β RaC
 IV → V
                    β
                ────────→ RaC ──α─→ RaD ──β─→ RaE ──β─→ RaF ──α─→ End
                99.97%   VI   IV   V   VI   IV

                    ? α
                ────────→ RaC₂ ──→ End
                0.03%   III   IV

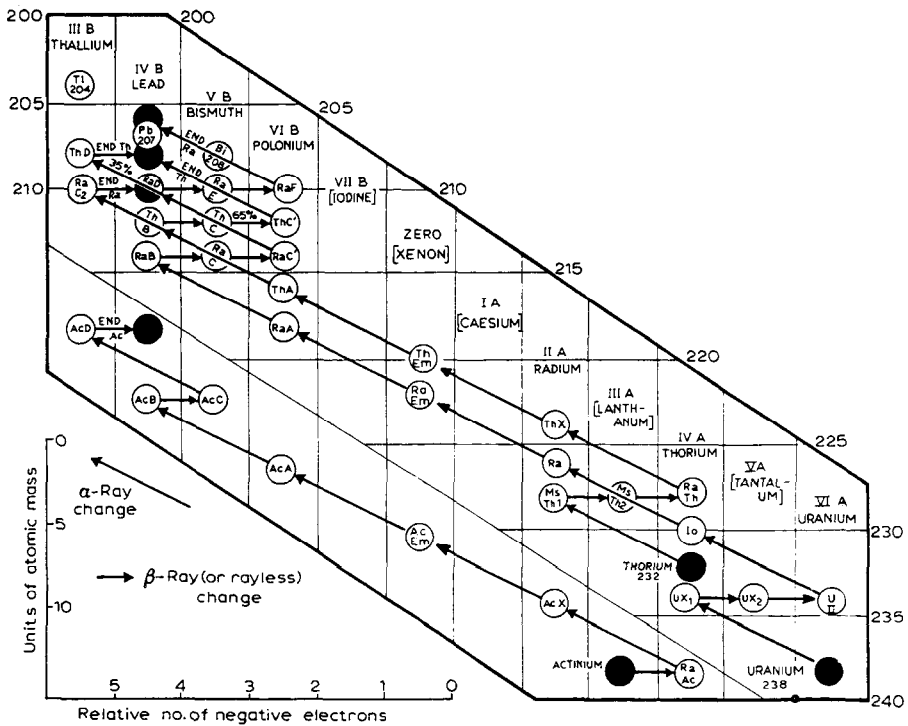*** The atomic weights shown for the actinium series are purely provisional(see p. 395).

Fig. I. Radio-elements and Periodic Law. All elements in the same place in the Periodic Table are chemically non-separable and (probably) spectroscopically indistinguishable.

in the *Chemical News* published simultaneously. At the time, in the absence of any special theory of atomic structure, or of the nature of positive electricity, this was natural, but now it is more in accord with our theoretical knowledge to represent the abscissae as units of positive electricity (Fig. 2). Either represents the fundamental fact that in the expulsion of an α-particle twopositive charges are lost by the atom, and in the expulsion of a β-particle one negative.

"Whenever, by the expulsion of α- or β-rays, two or more elements come to occupy the same place in the Periodic Table, then, independently of all other consideration, such as the atomic weight, the disintegration series to which the element belongs, its radioactive character and the nature of the radioactive changes, in which it is produced, or by which it is transformed, these elements occupying the same place, are non-separable from one another, and are, so far as is known, identical in chemical character." (1913, p. 263).

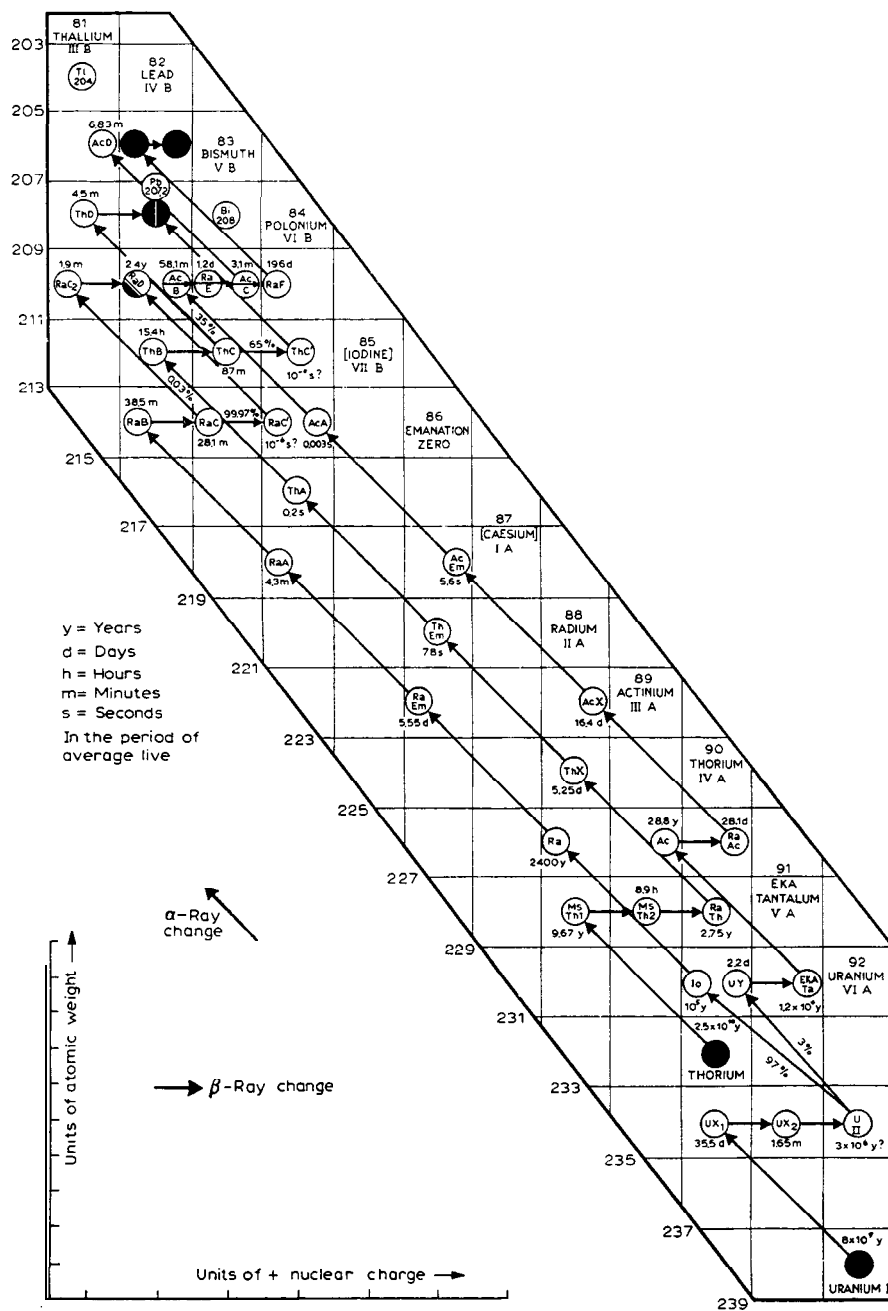Of the radio-elements still uncharacterized, the generalization shows that

Fig. 2. Radio-elements and Periodic Law. All elements in the same vertical column
are isotopes.

the A- and C-members must be non-separable from polonium and radium C_2, thorium D and actinium D from thallium. As regards radium A, thodrium D and actinium D these predictions were at once confirmed by Fleck[11].

The rules demanded that uranium X must consist of two successive products uranium X, and uranium X_2, the latter a new member in the vacant place in the Periodic Table, between uranium and thorium, and therefore analogous to tantalum. Fajans and Göhring proved this at once[12]. Uranium X_2 is very short-lived product responsible for the more penetrating of the two types of β-rays given by "uranium".

Since actinium is in the IIIrd family, its parent must be chemically identical with radium in the IInd family, if it is produced in a β–ray change, and in the Vth family, in the place occupied by uranium X_2, if it is produced in an α-ray change.

Lastly, and most important of all, the generalization showed that the ultimate products of all three series in all branches must be in the place occupied by the element lead. The atomic weight of the lead from both branches of the thorium series is 208, and that of the lead from uranium, in the main branch, 206. Common lead with the atomic weights 207.2 might well be a mixture of both.

In this same year, 1913, Sir Joseph Thomson and F. W. Aston announced the discovery by the positive-ray method of a constituent of atmospheric neon of atomic mass 22 in addition to the main constituent of atomic mass 20.

"The discovery is a most dramatic extenions of what had been found for elements at one extreme of the Periodic Table to the case of an element at the other extreme, and strengthens the view that the complexity of matter in general is greater than the periodic law alone reveals." (1913, p. 266).

The theoretical interpretation of isotopes

In 1911 Rutherford put forward, in a somewhat tentative form, his now well-known "nuclear theory" of atomic structure, to account for the scattering of α– and β–particles in their passage through matter. To account especially for the large angles through which occasional α–particles are deflected it was necessary to suppose that these were the result of single encounters between the α-particle and an atom struck. For this to be possible, there

must exist within the atom a much more powerful electric field than, for example, in the Thomson atom. Rutherford's supposed that, at the centre of the atom, there existed a nucleus of very minute dimensions relatively to the atomic volume, upon which was concentrated a large charge of one sign, the rest of the atom being occupied by a number of single charges of the opposite sign which neutralized the central or nuclear charge. At first nothing was postulated in the theory as to the sign of this central charge, whether positive or negative, or as to the constitution of the nucleus. But it was natural to regard the central charge as positive, and the rest of the atom to be occupied by a system of negative electrons. In the next two years, much further experimental evidence on scattering accumulated, and the theory began to assume a more definite form.

"Single scattering" on this theory should be proportional to the square of the nuclear charge. The experimental results indicated that it was approximately proportional to the square of the atomic weight of the scattering atom, and that there must be one unit of nuclear charge for two units of atomic weight, approximately. In 1911 Barkla[14] arrived at the same conclusion, as to the number of electrons in the atom from the scattering of X-rays.

In 1911, Van den Broek conceived the idea that "to each possible* intraatomic charge there corresponds a possible element". This necessitates that successive elements in the Periodic Table should differ by one unit of nuclear charge and by one electron in the outer shell, as was subsequently practically directly established by the periodic law generalization already considered.

To account for the supposed experimental result that there was one unit of charge for two units of atomic weight, Van den Broek at first attempted to revive the old and discarded "cubic" Periodic System of Mendeleeff, because it accommodated 120 elements. Since the last, uranium, has the atomic weight 240 it satisfies this requirement of giving on the average two units of mass per "place", i.e. per unit of charge. His attempts to fit the radioactive elements into this scheme appeared purely fanciful. But he then found that the actual experimental results for scattering were in entire accord with his own idea, on the accepted Periodic Table, which accomodates some 90 elements. Thus uranium in the last place with an intra-atomic charge of about 90, must have between 2 and 3 units of mass per unit of charge. So that, if its nucleus be imagined to be composed of 60 α-particles with charge 120, there must be present is also 30 electrons to give the nuclear charge 90.

* "Possible" here meant "integral"

This suggestion of Van den Broek was adopted by Bohr[16] in his theoretical researches on the structure of the atom. Bohr's views required that the electronic system is stable, so that to remove an electron involves the expenditure of energy. Hence it followed that the B-particles expelled in radioactive change must come from the nucleus and not from the external electronic system.

I had arrived at the same conclusion from totally different evidence[17]. If, for example, the two electrons that are expelled as B-rays, when uranium X, changes into uranium II, come from the same region of the atom as the two that are lost when U^{IV} in uranous salts is oxidized to U^{VI} in uranic salts, then the latter ought to be chemically non-separable from thorium, just as uranium X_1 is. Fleck[18], trying this, found that, whereas there is a very close resemblance between uranium in the uranous salts and thorium, yet the two may be separated chemically without difficulty.

The expulsion of two + charges as an α-particle and of two electrons as β–particles from the nucleus causes the element to come back to the original place that it occupied in the Periodic Table. It followed therefore that the place in the Periodic Table is an expression of the *nett* nuclear charge, i.e. of the difference between the numbers of positive and negative charges in the nucleus. Thus the chemically identical elements - or isotopes, as I called them for the first time in this letter to *Nature,* because they occupy *the same place* in the Periodic Table - are elements with the same algebraic or nett nuclear charge, but with different numbers of + and - charges in the nucleus. On the view that the concentrated positive charge is the massive particle in the atomic structure, since positive electricity has never been observed free possessing less than the mass of an atom, the atomic weight of the isotope is a function of the *total* number of positive charges in the nucleus and the chemical character a function of the *nett* number.

Though the nucleus possesses electrons there can be no in- or out-going of electrons between the nucleus and the external electronic system. Thus Rutherford's atom affords for the first time a clear picture of the difference between a transmutational (radioactive) change and chemical one. Changes of the number of electrons in the external system are chemical in character and produce changes in the valency of the element. These are reversible and have no effect at all on the central nucleus. Whereas changes of the nucleus are transmutational and irreversible, and they instantly impress changes upon the external electronic system to make it conform to the new nucleus. So far as I was concerned, this interpretation of isotopes, in the light of Van den

Broek's conception and Rutherford's nuclear atom, resulted in a great clari-
fication of my own ideas. It was completely independent of Moseley's
researches into the spectra of X-rays, which then had not been published.

Moseley[19] determined by the crystal reflection method the wavelengths
of the homogeneous secondary X-rays, characteristic of the chemical ele-
ments, discovered by Barkla. He found that the squares of these wavelengths
are a function of a number, now called atomic number, which alters by one
unit in passing from the X-ray spectrum of any element to that of the next
to it in the Periodic Table. He was thus able to extend the definite determina-
tion of the number of places in the Periodic Table, which had been accom-
plished for the elements from uranium to thallium by radioactive change,
to the rest of the Periodic Table as far as aluminium. Moseley's atomic
number is the same as Van den Broek's intra-atomic charge and represents(I)
the nett positive charge on the nucleus, (2) the number of electrons in the
outer electronic system of the atom, (3) the number of the place in the Period-
ic Table occupied by the element, counting from hydrogen as unity to
uranium as 92, as was first definitely determined later by his method.

The origin of actinium

In conclusion, I may very briefly deal with the verification of the two chief
predictions from the α–and β–change generalization.

The possibility that actinium resulted in a β–ray change necessitates that
its parent should be an isotope of radium.This I disproved directly by an exam-
ination of a specimen of Giesel's radium bromide that had been kept ten
years without any chemical treatment[20]. There was a total absence of radio-
actinium, after that time, in the preparation. The remaining alternative that
actinium was produced in an α-ray change from "eka-tantalum", a missing
element occupying the place between uranium and thorium, and isotopic
therefore with uranium X_2, was finally established by the independent work
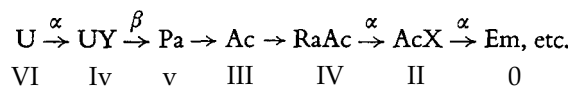of Hahn and Meitner[21], and of J. A. Cranston and myself[22], in 1918.

As regards our work, we found that uranium X_2 could be readily vola-
tilized from uranium X_1 in a stream of carbon tetrachloride and air at an
incipient red-heat. Thus we used uranium X_2 as a radioactive indicator of
the chemical properties of the unknown substance, something in the same
manner first employed by Paneth and von Hevesy. When pitchblende was
treated in the same way, a sublimate was obtained free initially from all the

known pm-emanation members, but continuously generating actinium, as shown by its characteristic emanation, with the lapse of time.

Hahn and Meitner found the missing element in the insoluble residues from the treatment of uranium minerals for radium, and separated it by methods designed to remove tantalum, which was added in minute quantity as required to assist the separation. From their work, the new element, protoactinium as they call it, promises to be of more than ordinary interest. They have not succeeded in separating it from the tantalum present or added during the operations. It gives α– ray of low range, generates actinium at a rate which has enabled the average life of the latter substance at length to be definitely established, as 28.8 years. The production of protoactinium from uranium has been demonstrated by the examination of old uranium salts. From the quantity present, its average life is 17,000 years. (Mr. Mennie working in my laboratory, with a large quantity of uranium, which I had very carefully purified in 1909, has confirmed this.) This period corresponds with a quantity of 72 mg of protoactinium per 1,000 kilograms of uranium, or 333 mg of radium, in minerals. So that it appears likely that it will be the second new radio-element to have its atomic weight, spectrum and chemical nature determined, in the same way as Mme. Curie accomplished for radium.

Protoactinium appears to be produced as a branch product, claiming 3% of the total number of atoms disintegrating, from either uranium I or uranium II through Antonoff's uranium Y, which is its direct parent. But it has also been suggested by Piccard that it may be derived from a totally independent isotope of uranium (actinouranium). The series runs:

$$\text{U} \xrightarrow{\alpha} \text{UY} \xrightarrow{\beta} \text{Pa} \rightarrow \text{Ac} \rightarrow \text{RaAc} \xrightarrow{\alpha} \text{AcX} \xrightarrow{\alpha} \text{Em, etc.}$$
$$\quad \text{VI} \qquad \text{Iv} \qquad \text{v} \qquad \text{III} \qquad \text{IV} \qquad \text{II} \qquad 0$$

The atomic weight of actinium is thus still indefinite.

The atomic weight and spectrum of lead and ionium

The ultimate product:. Immediately after the prediction that the main end products of uranium and thorium must be different isotopes of lead with atomic weights 206 and 208, determinations were undertaken of the atomic weight of lead separated from thorium minerals as free as possible from uranium, and of that from uranium minerals as free as possible from thorium.

These resulted in the complete verification of the prediction. The highest value yet found for "thorium-lead" is 207.9 (Fajans and Hönigschmid) and the lowest value for "uranium-lead" 206.05, the accepted value for common lead being 207.20.

In my own work, 30 kilograms of Ceylon thorite, containing 55% of thorium, about 1% of uranium and 0.4% of lead, was hand-sorted, and, from 20 kilograms of selected pieces, 80 grams of metallic lead were separated.

The view that isotopes have identical external electronic systems naturally leads to the deduction that their atomic volumes must be identical and, therefore, that the atomic weights must be proportional to their density. The thorite lead was cast in vacuo and its density compared with that of a similarly purified and treated specimen of common lead. Its density was found to be 0.246% greater, leading to a calculated value of 207.7 for the atomic weight. By fractional distillation in vacuo three fractions were obtained and the atomic weight of the middle one found, from the ratio $Pb : PbCl_2$ to be 207.69, as compared with 207.20, found in a parallel estimation of ordinary lead. The war interrupted these researches, but it is of interest to record that, for another specimen of my thorite lead, Hönigschmid found, by the silver titration method, 207.77 ± 0.014[23].

Simultaneously, work on lead from uranium minerals by T. W. Richards and his students at Harvard, and by Hönigschmid and Mlle. Horovitz, gave values all below the international figure. In the case of the two most carefully selected minerals, Morogoro uraninite and Norwegian bröggerite, values 206.46 and 206.063 were found[24]. For these also the atomic volume was found to be the same as for ordinary lead, the density being as much less as the atomic weight is less than for ordinary lead. For a similar reason, that the molar solubilities of isotopes must be equal, the actual solubilities of salts of different isotopes must be proportional to their molecular weights. Two independent determinations of the melting point of the lead of radioactive origin failed to reveal any difference from that of ordinary lead. To ordinary methods the spectrum of the lead prepared from radioactive minerals is identical with that of common lead.

Hönigschmid and Horovitz have determined the atomic weight of thorium and compared it with that found by the same methods for the ionium-thorium preparation prepared by Auer von Welsbach. For the first they obtained the value 232.12, and for the second 231.51, a difference of atomic weight of 0.61, corresponding with a percentage of ionium of about 30, if

230 be the value for the latter. This is in conformity with the period of ionium and the a-radiation of the preparation. But the spectrum of the iomum-thorium preparation was in every respect identical with that of thorium.

Although no difference in spectrum is observable by ordinary methods, Harkins and Aronberg[25], photographing the strongest line of the lead spectrum, 4058, in the sixth order of a 25-cm Rowland grating, found a minute difference. The wavelength of the line for uranium lead of atomic weight 206.34 is 0.0043 Å greater than that for common lead. This infinitesimal difference has been confirmed by Merton[26], who found also for my thorite lead a wavelength 0.0022 Å less than that of common lead.

Summary

We may now sum up the various distinct steps in this long and tangled story of the origins of the conception and discovery of isotopes.

I. Experimental methods are available, uniquely for the radio-elements, which enable isotopes to be severally recognized, by a suitable combination of chemical analyses at appropriate intervals of time, whereby, owing to the successive changes of the constituents, they may be separated, although chemical analysis alone is quite unable to effect this separation. This dates from 1905.

2. The complete chemical identity of isotopes, as distinct from close chemical similarity, came gradually to be recognized. McCoy and Ross were the first to express a definite opinion in this sense (1907).

3. The existence of chemical identities among the radio-elements led to the deduction that they might exist among the common elements and be responsible for the exceptions in the Periodic classification, and for the fact that the atomic weights in some cases depart widely from integral values. Strömholm and Svedberg first made this deduction (1909).

4. The recognition of the effect of the expulsion of, first, the a- and then, the β-particle (1911 and 1913) led to the correct placing of all three disintegration series from end to end in the Periodic Table. On the experimental side the names of A. Fleck, and on the theoretical side that of G. von Hevesy and A. S. Russell, but pre-eminently that of Kasimir Fajans, are associated with this advance.

5. The identity of isotopes was extended to include their electrochemistry

(Paneth and Hevesy) and their spectra (Russell and Rossi), though here infinitesimal differences were subsequently found (Harkins and Aronberg, Merton).

6. Isotopes, on Rutherford's theory of atomic structure, are elements with identical external electronic systems, with identical nett positive charge on the nucleus, but with nuclei in which the total number of positive and negative charges and therefore the mass is different. The originator of the view that the places in the Periodic Table correspond with unit difference of intra-atomic charge is Van den Broek.

7. Moseley extended this view to the non-radioactive elements, and ultimately for the whole Periodic Table, and the definite determination of the number and sequence of the places in it became possible.

8. The chemistry of the radioactive elements and the lacunae previously existing in the radioactive series, especially in connection with the origin of actinium, have been cleared up, and this led to the discovery of a new element, proto-actinium (eka-tantalum) in uranium minerals, occupying the place between uranium and thorium and existing in sufficient quantity for the compounds of the element to be prepared in a pure state, and its spectrum and atomic weight ultimately to be determined, as Mme. Curie did for radium. Cranston and I share with Hahn and Meitner the original discovery, but the subsequent developments are due to the latter.

9. The preparation from radioactive minerals of different isotopes of lead followed and the determination of their atomic weight, spectrum, density and other properties, established that the same chemical character and atomic volume can coexist with differences of atomic weight. The work on the ionium-thorium mixture from pitchblende is a second example.

10. The last result, and perhaps the most important of all, is the subject of the award of the Nobel Prize for Chemistry for 1922.

1. H. N. McCoy and W. H. Ross, *J. Am. Chem. Soc.,* 29 (1907) 1709.
2. B. Keetman, *Jahrb. Radioaktiv.,* 6 (1909) 268.
3. A. Schuster, *Nature,* 91 (1913) 30, 57.
4. A. Fleck, *Chem. News,* 106 (1912) 128.
5. A. Fleck, *Proc.Ckem. Soc.,* 29 (Jan. 30, 1913) 7; *Trans. Chem. Soc,* 103 (1913) 381.
6. G.v. Hevesy, *Physik.Z.,* 14 (Jan.15, 1913) 49.
7. G.v. Hevesy, *Phys.Z.* 14 (Jan. 15, 1913) 60.
8. A.S. Russell, *Chem.News,* 107(Jan.31,1913)49.

9. K. Fajans, *Physik.Z.,* 14 (Feb. 15, 1913) 131.

10. F. Soddy, *Chem. News,* 107 (Feb. 28, 1913) 97; *Jahrb. Radioaktiv.,* 10(1913) 188.

11. A. Fleck, *Trans. Chem. Soc.,* 103 (1913) 1052, compare also W. Metzener. *Ber.,* 46 (1913) 979.

12. K. Fajans and O. Göhring, *Physik.Z.,* 14 (1913) 877.

13. E. Rutherford, *Phil. Mag.,* [6] 21 (1911) 669.

14. C. G. Barkla, *Phil. Mag.,* [6] 21 (1911) 648.

15. A. van den Broek, *Nature,* 87 (1911) *78;* 92 (1913) 372.

16. N. Bohr, *Phil Mag.,* [6] 26 (1913) 476.

17. F. Soddy, *Nature,* 92 (1913) 400.

18. A. Fleck, *Trans. Chem. Soc.,* 105 (1914) 247.

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