

## RADIOACTIVITY.

IN the last section of the 1910 Report, "Chemical Relationships of the Radio-elements," the existence of groups of radio-elements possessing identical chemical properties was shown to foreshadow "some embracing generalisation which will throw light, not only on radioactive processes, but on the elements in general and the Periodic Law."<sup>1</sup> In 1911 the first step in this direction was made, when it was recognised that the expulsion of the  $\alpha$ -particle causes the radio-element to change its position in the periodic table, not into the next family, but into the next but one in the direction of diminishing group number and diminishing atomic mass.<sup>2</sup> Last year doubtful points in the sequence of changes, consequent upon the branching of the disintegration series at the *C*-members, and on the existence, in uranium, of two chemically identical elements, uranium-*I* and -*II*, were cleared up, and the important step made that the *B*- and *C*-members of the three series exhibit identical electrochemical behaviour.<sup>3</sup> In the meantime, a systematic study of the chemical nature of those disintegration products not hitherto thoroughly studied from a chemical point of view had resulted in a remarkable extension of the feature which dominates the chemistry of the radio-elements.<sup>4</sup> Radioactinium was shown to be chemically identical with thorium; mesothorium-*II* with actinium; the three *B*-members and radium-*D* with lead; the three *C*-members and radium-*E* with bismuth; thorium-*D* and actinium-*D* with thallium<sup>5</sup>; and radium-*A* with polonium. Thus, not a single one of the radio-elements, known at the commencement of the year,

<sup>1</sup> *Ann. Report*, 1910, 285.

<sup>2</sup> F. Soddy, "Chemistry of the Radio-elements," 1911, p. 30.

<sup>3</sup> *Ann. Report*, 1912, 311, 321, 319. Compare also E. Marsden and R. H. Wilson, *Phil. Mag.*, 1913, [vi], 26, 354; *A.*, ii, 907; P. Beer and K. Fajans, *Physikal. Zeitsch.*, 1913, 14, 947; *A.*, ii, 907; A. B. Wood, *Phil. Mag.*, 1913, [vi], 26, 586; *A.*, ii, 908; K. Fajans, *Physikal. Zeitsch.*, 1913, 14, 951; *A.*, ii, 903; G. von Hevesy and L. von Putnok, *Physikal. Zeitsch.*, 1913, 14, 63; *Phil. Mag.*, 1913, [vi], 25, 415; *A.*, ii, 175.

<sup>4</sup> A. Fleck, *Chem. News*, 1912, 106, 128; 1913, 107, 95; *T.*, 1913, 103, 381, 1052.

<sup>5</sup> Compare also W. Metzener, *Ber.*, 1913, 46, 979; *A.*, ii, 375.

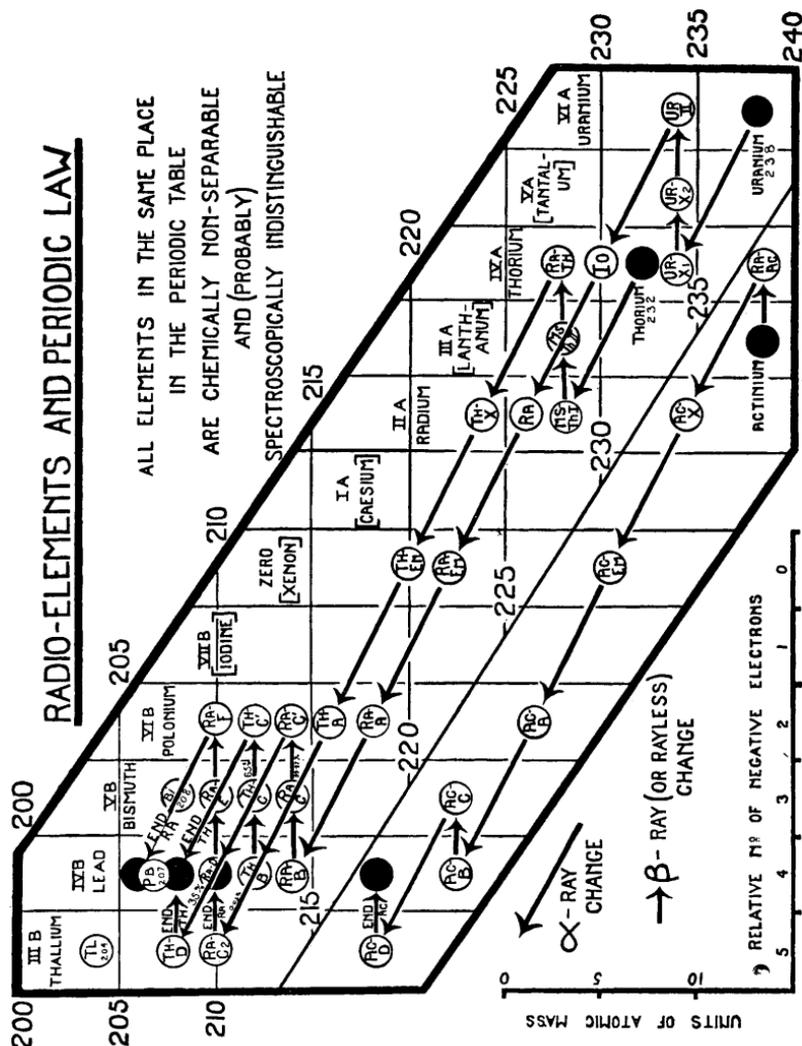
has a peculiar chemical nature unshared by others. All are chemically indistinguishable from one or other of the elements occupying the last twelve places of the periodic table, from thallium to uranium. With the sequence of changes fully elucidated and the chemical character of the majority of the radio-elements established, the  $\alpha$ -ray rule was shown to hold generally, and, equally generally, a similar rule for the  $\beta$ -ray changes was found to apply. In the  $\beta$ -ray change, the element shifts its position in the periodic table in the opposite direction to that in the  $\alpha$ -ray change, but into the next family, not into the next but one.<sup>6</sup> These two simple rules, consistently applied to the three disintegration series, constitute a sweeping generalisation connecting the chemical character of the radio-element, and the position it occupies in the periodic table with the kind of radioactive change in which it is produced. In addition to the purely chemical discoveries considered, an electrochemical examination of the radio-elements led independently to the same generalisation. It was found that the expulsion of the  $\alpha$ -particle resulted in a product more electro-positive, and of a  $\beta$ -particle more electro-negative, than the parent.

The generalisation is illustrated in the chart (Fig. 1). This satisfactorily accounts for all the peculiar features that characterise the chemistry of the radio-elements. Whenever, by the expulsion of  $\alpha$ - or  $\beta$ -rays, two or more elements come to occupy the same place in the periodic table, then, independently of all other considerations, 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. Each vertical row of the diagram consists of such a group of chemically identical elements. The ten occupied places contain over forty distinct elements, whereas if chemical analysis alone had been available for their separate recognition, only ten elements could have been distinguished. The places at the end of the periodic table, and probably elsewhere in the table, thus represent, not single homogeneous elements as has hitherto been supposed, but groups of elements identical in chemical character. To express this newly discovered complexity of matter, the terms "isotopic elements" or "isotopes" have been coined. Thus radiothorium, ionium, thorium, uranium- $X_1$ , and radioactinium

<sup>6</sup> A. S. Russell, *Chem. News*, 1913, **107**, 49; *A.*, ii, 274; K. Fajans, *Physikal. Zeitsch.*, 1913, **14**, 131, 136; *Ber.*, 1913, **46**, 422; *A.*, ii, 276, 277; *Ber. Deut. physikal. Ges.*, 1913, **15**, 240; *A.*, ii, 493; *Le Radium*, 1913, **10**, 171; *A.*, ii, 660; F. Soddy, *Chem. News*, 1913, **107**, 97; *Jahrb. Radioaktiv. Elektronik*, 1913, **10**, 188; *A.*, ii, 275.

are a group of isotopic elements, the calculated atomic masses of which vary from 228 to 234. They all occupy the same place in

Fig. 1.



Reproduced by the courtesy of the British Association from a block illustrating "The Radio-elements and the Periodic Law," by F. Soddy, Section B, Birmingham, 1913.

the periodic table, and are chemically indistinguishable. This material identity, however, extends far beyond the chemical pro-

perties in the narrow sense, and embraces probably nearly all the common physical properties also, so that the experimental means capable of distinguishing and separating isotopes are very limited.<sup>7</sup> Thus, eleven years ago, the result that the radium and thorium emanation condense at practically the same temperature seemed very extraordinary. Now there is every reason to believe that isotopes will prove to be indistinguishable in volatility no less than in chemical properties. The question whether isotopes have the same spectrum, for example, was discussed for the case of ionium and thorium last year.<sup>8</sup> Important new evidence can be urged both for and against the general view. The recent generalisation that the magnitude of the atomic weight enters exactly into the series relationships of spectra, and the expectation that has been advanced that, ultimately, it should be possible to determine atomic weights from these series relationships more accurately than by chemical analysis,<sup>9</sup> is obviously opposed to the possibility that elements of different atomic masses can have the same spectra.

*Neon and Metaneon.*—On the other hand, what appears to be a case of isotopic elements outside the radioactive sequences has been discovered. As has been remarked, very few material properties depend directly on atomic mass. Fractional diffusion of gases is almost the only property that can be expected to effect a partial separation of a group of isotopic elements into their constituents. Whilst, to detect the non-homogeneity if it exists, the new positive ray method of Sir J. J. Thomson<sup>10</sup> is again almost the only one available. The examination of atmospheric neon by this method revealed the presence of atoms, in relatively small proportion, of mass 22, in addition to the known atom of mass 20. The relative proportion of the two kinds of atoms was unchanged after a prolonged fractionation of the gas by cold charcoal; but fractional diffusion showed that atmospheric neon is not homogeneous, and a partial separation, attested by a change of density, was effected by this means. No change in the spectrum corresponding with the change of density was observed, however, and the two elements appeared to be identical in all properties, except atomic weight.<sup>11</sup>

This accords with what has been found in the case of ionium

<sup>7</sup> "Chemistry of the Radio-elements. Part II. The Radio-elements and the Periodic Law." By F. Soddy: Longmans, Green & Co., 1914.

<sup>8</sup> *Ann. Report*, 1912, 321.

<sup>9</sup> W. M. Hicks, *Phil. Trans.*, 1913, A, **213**, 323; *A.*, ii, 810.

<sup>10</sup> Sir J. J. Thomson, Bakerian Lecture, *Proc. Roy. Soc.*, 1913, **89**, A, 1; *A.*, ii, 820.

<sup>11</sup> F. W. Ashton: Paper communicated to the British Association, Section A., Birmingham, 1913.

and thorium, as regards the spectra, and in the case of the radium and thorium emanation, as regards the volatility, and indicates that isotopes will prove to be identical in these respects as they are in chemical character. At the same time, the discovery is a most dramatic extension of what has been found for the 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. Although the complexity is greater, the problem of atomic structure has been much simplified, because the generalisation gives a probable explanation of the absence of exact simple numerical relations among the atomic weights.

*Uranium- $X_2$  or Brevium.*—When the rules were first applied there was one obvious lacuna between uranium- $X$  and uranium- $II$ , for, by the rules, two successive  $\beta$ -ray changes are necessary to shift an element in group IV into group VI. This suggested that uranium- $X$  was probably complex, and that its two types of  $\beta$ -rays, one very hard and the other very soft, result in two successive changes, with the intervention of an intermediate element occupying the place in group V, hitherto vacant. The new element would be the homologue of tantalum. The resolution of uranium- $X$  into two distinct radio-elements, named uranium- $X_1$  and uranium- $X_2$ , followed almost immediately.<sup>12</sup> By application of the electro-chemical rules, it was to be expected that uranium- $X_2$  would be more easy to separate electro-chemically than uranium- $X_1$ , and it was found that polished lead plates, immersed for a minute in a feebly acid solution of uranium- $X$ , gave the hard  $\beta$ -rays of uranium- $X$ , which decayed with a half-period of 1.15 minutes. This corresponds with a period of average life for uranium- $X_2$  of 100 seconds, whereas the period of uranium- $X_1$ , the parent, is 35.5 days. The chemical analogy of the new member to tantalum is clearly shown by other methods of separation. If uranium- $X$ , separated from uranium, together with thorium, in acid solution, is added to an alkaline solution of potassium hexatantalate, uranium- $X_2$  is almost quantitatively precipitated with the tantalic acid, whilst uranium- $X_1$  remains in solution with the thorium. Similarly, the separation may be effected by filtering the uranium- $X$  solution through a layer of moist tantalic acid. By this and many other reactions it was proved that uranium- $X_1$ , of average period 35.5 days, emits only the soft  $\beta$ -rays and produces uranium- $X_2$ , of average period about 1.6 minutes, which emits the hard  $\beta$ -rays.

<sup>12</sup> K. Fajans and O. Göhring, *Naturwissenschaften*, 1913, **1**, 339; *Physikal. Zeitsch.*, 1913, **14**, 877; *A.*, ii, 909; O. Hahn and L. Meitner, *ibid.*, 758; *A.*, ii, 821; A. Fleck, *Phil. Mag.*, 1913, [vi], **26**, 528.

Although uranium- $X_2$  is chemically analogous to tantalum, it should prove to be separable from it, just as radium is from barium, or polonium is from tellurium. As it is a new chemical type, its discoverers have given it the distinctive name "Brevium." At present it is unique among the radio-elements, in occupying a place in the periodic table by itself. This distinction is, however, likely to be only temporary, as already it is necessary to postulate as isotope to account for the origin of actinium.

*Origin of Actinium.*—The unsolved problem of the origin of actinium has been considerably narrowed down by the periodic law generalisation. Being in group III, its parent must be in group V, isotopic with brevium, if it is formed in an  $\alpha$ -ray change, or in group II, isotopic with radium, if it is formed in a  $\beta$ -ray change. As radium is the only member of the uranium series in group II, the latter alternative is equivalent to supposing that radium itself is the parent—that, in addition to the  $\alpha$ -ray change into the emanation, radium suffers a branch  $\beta$ -ray change into actinium. Although this is supported by the fact that radium, and also thorium-X, in the same place, give, in addition to  $\alpha$ -rays, some soft  $\beta$ -rays, which constitute something of an exception to the simple rules, the production of actinium from radium has been disproved.<sup>13</sup> No detectable amount of actinium was found in a preparation of radium bromide which had been kept for ten years without interference, and contained 13.2 mg. of radium (element). Were actinium the direct product of radium, this negative result would mean that the period of actinium must be at least fifteen million years, whereas it is practically certain that its period cannot be as great as one hundred years.

The alternative is to suppose that actinium results from an element in group V by an  $\alpha$ -ray change, and, since brevium is short-lived and gives no  $\alpha$ -rays, this requires that an unknown isotope must exist in the uranium series. The suggestion has been made that the series may branch at uranium- $X_1$ ,  $\beta$ -rays being expelled in both branches, so that two isotopic products result.<sup>14</sup> The branchings so far studied have been of the character that in one mode a  $\beta$ -ray change is followed by an  $\alpha$ -ray change, and, in the other mode, an  $\alpha$ -ray change is followed by a  $\beta$ -ray change. The suggestion that in both branches the same type of ray is expelled is novel, and although not necessarily improbable, would, if adopted, largely increase the number of possibilities to be taken into account. One, in particular, seems worth discussion.

*Uranium-Y.*—In 1911 Antonoff found that the soft  $\beta$ -radiation

<sup>13</sup> F. Soddy, *Nature*, 1913, **91**, 634.

<sup>14</sup> O. Hahn and L. Meitner, *Physikal. Zeitsch.*, 1913, **14**, 752; *A.*, ii, 821.



experimentally indistinguishable, save by indirect methods, such as the determination of the atomic weight of actinium and its products. The scheme shown would make the atomic weight of actinium 230, whereas the alternative would make it 226, identical with that of radium.

A generalisation in this field favours the latter alternative.<sup>18</sup> As a rule, in any group of isotopes, the  $\alpha$ -ray giving members are the more stable the higher the atomic weights, and the  $\beta$ -ray giving members are the more stable the lower the atomic weight. Thus, in the thorium group, the order of increasing stability and atomic weight among the  $\alpha$ -ray members is: radiothorium (2.9 years, 228), ionium (100,000 years, 230), thorium ( $4 \times 10^{10}$  years, 232). Since radioactinium is the least stable of all in this group, its atomic weight ought to be less than 228. From this generalisation the atomic weights of actinium and its products generally appear to be similar to that of radium and its products. The generalisation does not hold without exception, the most notable being polonium, which at once is the most stable and has the smallest atomic mass of the six  $\alpha$ -ray giving members of this isotopic group.

*Nature of the End-Products.*—The connexion between the changes and the properties of the products enables the place occupied, and the whole chemical character of those members, to be predicted without uncertainty, in the case where, either by reason of excessive instability—the *A*- and *C'*-members—or excessive stability—the end-products—the elements are outside the range of experimental investigation. Hitherto there has been a general belief that the end product of radium was lead, but there was no evidence whatever as to the nature of the end-products of actinium and thorium. Fig. 1 shows that *all* the five end-products fall into the same place, and must be the isotopes of lead. The calculated atomic weights of the two end-products of the thorium series are the same, namely, about 208, and that of the main end-product of uranium is 206, whereas in the international list the atomic weight of lead figures as 207.1. This raises directly the question of the homogeneity of lead, for if it were a mixture of the end-products of uranium and thorium in similar proportion, the atomic weight would be about that actually found. On the other hand, if the generalisation of Fajans holds good, it is to be expected that the stability of these various isotopes will be connected with their atomic weight. If, for example, the two supposed end-products of the thorium series, like the corresponding product, radium-*D*, expelled a  $\beta$ -particle in further very slow changes, the product

<sup>18</sup> K. Fajans, *Le Radium*, 1913, 10, 171; *A.*, ii, 660; *Physikal. Zeitsch.*, 1913, 14, 951; *A.*, ii, 908.

would be, in chemical character and atomic weight, the same as bismuth; but as in the formation of these two products from the original parent substances different energies have been lost, the two isotopic bismuths so formed would not be identical, and one of them might further expel an  $\alpha$ -particle to give thallium (204), for both bismuth and thallium occur in radioactive minerals. This, of course, is a pure speculation. The interesting point is that, in stability, the supposed end-products of the thorium series should be intermediate between radium-*D* and radium-*G*, and, therefore, might not be sufficiently stable to accumulate in minerals and to constitute a perceptible fraction of the lead occurring therein. So that, although the atomic weight of lead, from radioactive sources as different as possible in regard to uranium and thorium content and geological horizon, should be re-examined, it by no means necessarily follows that any difference in atomic weight will be found.

*Branching of the Actinium Series.*—The earlier observation, which made it appear that actinium-*C* gave out two types of  $\alpha$ -rays, like thorium-*C*, but unlike in possessing ranges nearly the same,<sup>19</sup> has not been confirmed. In Fig. 1 the actinium series is represented as unbranched at the *C*-member. Without prejudice to this point, it appears certain that, if not the whole, at least very nearly the whole, of the actinium-*C* atoms disintegrate by the  $\alpha$ -ray mode, and what is, in this series, overwhelmingly the main branch is the minor branch in both the radium and thorium series. Recently some evidence of a branching has been obtained, precisely analogous to that which occurs in the other two series.<sup>20</sup>

In addition to the  $\alpha$ -rays, from actinium-*C*, of range 5.12 cm. in air at N.T.P., about one in 600 have been found of longer range, 6.1 cm. This is very nearly the range of the  $\alpha$ -rays of actinium-*A*. It must be remembered that recoiled actinium-*X* is always present in the active deposit of actinium, as ordinarily prepared,<sup>21</sup> so that further discussion may be postponed.

#### *The Structure of the Atom.*

On Rutherford's theory of atomic structure,<sup>22</sup> which was put forward to account for "single scattering"—that is, for the large angles through which a very small proportion of the  $\alpha$ -particles are deflected in their passage through matter, by rare, exceptionally close, single encounters with atoms—the mass of the atom, in association with positive electricity, about one unit of

<sup>19</sup> *Ann. Report*, 1909, 236.

<sup>20</sup> E. Marsden and R. H. Wilson, *Nature*, 1913, **92**, 29.

<sup>21</sup> *Ann. Report*, 1906, 363; 1907, 328; 1909, 246.

<sup>22</sup> *Ibid.*, 1911, 273.

charge per two units of mass, occupies a single central nucleus of excessively minute dimensions, in diameter only one ten-thousandth of the atomic diameter. Around this nucleus a number of negative electrons, equal to the value of the positive nuclear charge, circulate as an outer ring or shell. This atom is, of course, not stable according to ordinary electro-dynamical laws, for nothing apparently operates to prevent the dispersion of the extremely concentrated central positive charge, but it is now recognised that these laws require modification. The model has been used with very considerable success, in conjunction with Planck's theory of quanta, and leads to results, in connexion, for example, with the series relationships of the hydrogen and helium spectra, in striking accord with experimental determinations.<sup>23</sup>

It now appears that the magnitude of this central positive charge, in terms of the unit atomic charge, for example, that carried by the hydrogen ion, is probably the same as what is conveniently termed the "atomic number."<sup>24</sup> The atomic number is the number of the place an element occupies in the periodic table, when the successive places from hydrogen to uranium are numbered in sequence, hydrogen being unity, helium two, lithium three, and so on. If the eleven known representatives of the rare-earth group, between cerium and tantalum, exclusive, are all that exist, the atomic number for uranium would be 89. As regards the relative value of the nuclear charge, and its variation by units in passing from one place to the next of the periodic table, Fig. 1 shows this clearly. The  $\alpha$ -particle carries two positive atomic charges, and its expulsion causes the element to move two places in the table, that is, diminishes the atomic number by two. The  $\beta$ -particle is an atom of negative electricity, and its expulsion causes the element to alter its place by one in the opposite direction, that is, causes the atomic number to increase by one, whereas the successive expulsion of two  $\beta$ - and one  $\alpha$ -particle in any order brings the element to the same place and with the same atomic number, therefore, as initially.

The only assumption made is that both the  $\alpha$ - and  $\beta$ -particles are expelled from the central nucleus, not from the outer ring. This is in accord with the view, everywhere accepted, that radioactive changes are much more fundamental in character than any other known changes, and concern a region of the atom not affected in ordinary chemical and physical changes. As, on the theory, the mass of the atom is concentrated in the nucleus, and the

<sup>23</sup> N. Bohr, *Phil. Mag.*, 1913, [vi], **26**, 1, 476, 857.

<sup>24</sup> A. van den Broek, *Nature*, 1913, **93**, 373, 476; F. Soddy, *ibid.*, 399, 452; E. Rutherford, *ibid.*, 423.

$\alpha$ -particle constitutes an appreciable part of the atomic mass, there can be little doubt that the  $\alpha$ -particles have their origin in the nucleus. With regard to the  $\beta$ -particles, something like a direct proof has been obtained. If the  $\beta$ -particles came from the outer ring and were the same electrons as are concerned in electrochemical changes of valency, the loss of two  $\beta$ -particles by uranium- $X_1$  in the formation of uranium- $II$  should be equivalent to the loss of two electrons by uranous salts in their electrochemical oxidation to uranyl salts, and uranium in uranous salts should be isotopic with thorium. Uranous and thorium salts are very similar in chemical character, but a recent examination shows that they are readily separable from one another by chemical methods.<sup>25</sup> Hence the positively charged nucleus of the atom must contain some negative electrons—at least six in the case of uranium to account for the six  $\beta$ -rays expelled during the disintegration—which are to be distinguished from the electrons in the outer shell. If the mass of the uranium atom were due entirely to potential  $\alpha$ -particles or the nuclei of helium atoms, for which unit charge corresponds with two units of mass, there would be about thirty nuclear electrons to give 89 as the atomic number, whereas if, as has been surmised, the hydrogen nucleus were a second constituent of the nuclear structure, more must be postulated, as in this case unit mass goes with unit charge. It is the difference between positive and negative charges which gives the atomic number or the position of the element in the periodic table. Isotopes are elements for which the algebraic sum of opposite charges is the same, but the arithmetical sum is different.

The surprising results in crystallography and chemistry following upon the discovery that the  $X$ -rays are regularly reflected by crystals<sup>26</sup> are discussed in the report on Mineralogical Chemistry. Equally remarkable are the results in the field under discussion, for the property has enabled the wave-length of the  $X$ -rays, which is the order of ten thousand times smaller than that of visible light, to be determined. Each element gives a homogeneous characteristic  $X$ -radiation, and the wave-length of this characteristic radiation, equally with the wave-lengths of the luminous spectrum, is connected, by Bohr's theory, with the magnitude of the nuclear charge. The series of metallic elements from calcium to zinc in the periodic table, for which the atomic numbers extend from 20 to 30, were all in turn, excepting scandium, made the anti-cathode of an  $X$ -ray tube, and the spectrum in each case was found, by reflection from a crystal surface, to consist of two lines only, one strong and one weak. The wave-length of the stronger

<sup>25</sup> A. Fleck, *T.*, 1914, **105**, 247.

<sup>26</sup> *Ann. Report*, 1912, 302.

lines corresponded with the atomic numbers 19 to 29, showing unit difference of nuclear charge from place to place. What will be one of the most interesting points to chemists is that the nuclear charge increases in the order: iron, cobalt, nickel, which is the order the elements should be placed in the periodic table, and not in the order of the atomic weights: iron, nickel, cobalt. This is a definite proof that the "exceptions" to the sequence of atomic weights in the periodic table are real, and not due to erroneous determinations.<sup>27</sup> Much remains, obviously, to be worked out in this new field, but the results attained are sufficient to demonstrate how many novel methods of surprising power are converging simultaneously on the outstanding problem of the structure of atoms.

*Behaviour of Radio-elements in Solution, and to Solvents.*

Advantage has been taken of the fact that none of the known cations differ widely in mobility, and that the diffusion coefficient, in the presence of great excess of anion, varies inversely as the valency, in order to determine the valency of the radio-elements from diffusion experiments.<sup>28</sup> The first series of determinations gave results for the valency in accord with the chemical character for those radio-elements which had already been placed in the periodic table. Radium-*E* and the *C*-members, however, appeared to act as bivalent, and the *B*-members as univalent, when first determined by this method, in apparent disagreement with the isotopism of these elements to bismuth and lead respectively, as shown by the chemical examination published at about the same time. Later a re-examination gave values in accordance with the chemical evidence.<sup>29</sup> It was shown that, for example, thorium-*B* in certain circumstances acts as a univalent ion in the same way as lead does in the ion  $\text{PbCl}^+$ .

The chemical identity of isotopes has been extended to include the electro-chemical behaviour. The decomposition voltage curves for thorium-*C* and radium-*E* are the same as that of bismuth, and for thorium-*B* as that of lead. In addition thorium-*B* may be deposited on the anode under the same conditions as lead in the form of peroxide.<sup>30</sup> By the use of these radioactive substances as indicators, the behaviour of lead and bismuth for electrode potentials lying above the decomposition voltage has been studied. In the electro-deposition of radio-elements, the electrode potential is of the first importance. The passage of the current, and hence the

<sup>27</sup> H. G. J. Moseley, *Phil. Mag.*, 1913, [vi], **26**, 1024.

<sup>28</sup> G. von Hevesy, *Physikal. Zeitsch.*, 1913, **14**, 49; *A.*, ii, 174.

<sup>29</sup> *Idem. ibid.*, 1913, **14**, 1202; *A.*, 1914, ii, 16.

<sup>30</sup> F. Paneth and G. von Hevesy, *Monatsh.*, 1913, **34**, 1593; *A.*, ii, 1009.

difference between anode and cathode are entirely secondary. In practical electrochemical separations of one radio-element from another the addition of a perceptible amount of the isotope of the element it is not desired to separate is recommended. For example, in separating the *B*-members from the *C*-members, addition of lead hinders the deposition of the former, and results in a purer preparation of the *C*-member. An extended attempt to alter the concentration of radium-*D* to lead in a mixture by fractionation was unsuccessful. About twenty different methods failed to affect the least change in concentration.<sup>31</sup> This identity of behaviour has been made use of in the determination of the solubility of the excessively insoluble compounds of lead, such as the chromate and sulphide. To the lead is added a quantity of radium-*D*, infinitesimal in amount, but, after the formation of the products radium-*E* and -*F*, possessing intense radioactivity. The activity of the evaporated saturated solution of the insoluble lead compound enables the amount of lead present to be estimated, though this is analytically undetectable. In this way, the solubility of lead chromate in water at 25° was found to be 0.012 milligram per litre, and of lead sulphide, 0.3 milligram in water and 0.15 milligram in a saturated solution of hydrogen sulphide. Similar experiments could be carried out with thorium, radium, or bismuth, using ionium, thorium-*X*, or radium-*E*, respectively, as indicators. The possibilities of the method are very numerous.<sup>32</sup>

Two lines of work indicate that the radio-elements, although present in such infinitesimal concentrations, often behave in solutions as colloids rather than as electrolytes. Thus polonium may be separated from lead, radium-*D* and -*E*, by dialysing the solution through animal membranes, or thin parchment paper. The polonium remains behind in the cell in a pure condition.<sup>33</sup>

This may be used to effect the separation, but on a large scale the process recommended is the crystallisation of the hot saturated solution of the radio-lead nitrate solution, and electrolysis of the mother liquor, after addition of a little bismuth, between platinum electrodes, with a cathode potential not exceeding 0.08 volt, or current strength 0.16 milliampere per sq. cm. The polonium is removed from the electrodes by volatilisation at 1000°. The vapour condenses preferably on palladium or platinum, especially the former, rather than on other metals or on quartz, and so may be concentrated.<sup>34</sup>

<sup>31</sup> F. Paneth and G. von Hevesy, *Monatsh.*, 1913, **34**, 1393; *A.*, ii, 1008.

<sup>32</sup> *Idem. ibid.*, 1913, **34**, 1401; *A.*, ii, 1075; and G. von Hevesy, British Association, paper read before Section B, Birmingham, 1913.

<sup>33</sup> F. Paneth, *Monatsh.*, 1913, **34**, 461; *A.*, ii, 273.

<sup>34</sup> F. Paneth and G. von Hevesy, *Monatsh.*, 1913, **34**, 1605; *A.*, ii, 1011.

In acid solution, all the radio-elements dialyse normally as crystalloids; in neutral solution the velocity of dialysis in the case of polonium and radium-*E*, and in ammoniacal solution in the case of thorium-*B* also, is very much reduced. The diffusion-coefficients in acid, neutral and alkaline solutions of these elements bear out the conclusions drawn from the results of dialysis. The change from crystalloid to colloid is accompanied by a change of sign of the charge carried by the particles, the colloidal particles now migrating to the anode in an electric field. In a condition where both forms of the same radio-element exist together in solution, the migration of the ions to the cathode is not prevented, but only hindered, by the insertion of a parchment membrane, but the migration of the colloid particles to the anode is almost completely prevented by the membrane. The size of the colloidal particle is probably very small, and it consists of an aggregation of but few atoms compared with ordinary colloidal particles. It is altogether beyond the range of the ultra-microscope. This is to be expected in view of the excessively attenuated concentrations. When it is remembered that in none of these cases is the solution a saturated one, and, even for the most insoluble, the solubility product of the ions is very far from being approached, it is clear that the aggregation cannot be due to purely physical causes, but that some unknown chemical or adsorption phenomena are involved. Zsigmondy has suggested that the colloidal particles may be formed by the adsorption of the radio-element by existing colloid particles, such as aluminium hydroxide or silica, derived from the materials of the containing vessels. This explanation necessitates the specific adsorption by the colloid particles of some of the radio-elements, and not of others. A closer examination of the adsorption phenomena of the radio-elements is therefore called for. Along these lines may be expected an explanation of the very inexplicable fact that the radio-elements show such definite analytical and chemical reactions at such infinitesimal concentrations.<sup>34a</sup>

In the other research, it was found that the ratio of cathode to anode activity, obtained by the electrolysis of water containing dissolved radium emanation, could be varied in the proportion of 40,000 to 1 in a manner impossible to account for if the products, radium-*A*, -*B*, and -*C* existed as ions, but completely in accord with predictions made from the theory of colloids. In pure water, the *A*-member is deposited on the anode, the *B*-member on the cathode, and the *C*-member on both electrodes, and the view taken is that the *A*-member exists as a positive hydrosol, the *B*-member

<sup>34a</sup> F. Paneth, *Kolloid. Zeitsch.*, 1913, **13**, 1, 297; *A.*, ii, 747; 1914, ii, 19; K. Fajans and P. Beer, *Ber.*, 1913, **46**, 3486; *A.*, ii, 1010.

as a negative hydrosol, and the *C*-member in both forms. The addition of acids containing hydrion, or much more powerfully, of a multivalent cation such as that of aluminium, stabilises the positive hydrosol, whilst it neutralises and ultimately reverses the sign of the negative hydrosol. Basic ions, such as hydroxyl, or the citric acid anion, act in the opposite manner. In this way the deposition on either electrode may be suppressed, and that on the other accentuated. The transport of the products by the current is regarded as being due to electrophoresis rather than to electrolysis.<sup>35</sup>

An interesting series of experiments on the solubility of the active deposit of radium deposited from the emanation on surfaces of glass and metals, showed that from a glass surface only one-half of the active deposit is removed by boiling with acids, whereas with gold and platinum surfaces the soluble part is greater.<sup>36</sup>

There is little doubt that the recoil of the atom of radium-*B* in its production from radium-*A* must drive one-half of it to a perceptible depth beneath the surface, where it is protected from the attack of solvents. With radium-*B* deposited on a second plate, by recoil from the first, the whole is soluble. In organic solvents, especially carbon disulphide, radium-*A* is soluble, whereas the other two members are nearly insoluble. This is in accord with the position of radium-*A* in the sulphur family. When the active deposit is formed on plates of gold and platinum, which have been previously saturated electrolytically with hydrogen and oxygen respectively, it was found that oxygen diminishes the solubility, especially of radium-*C*, whilst hydrogen diminishes that of radium-*A*. It will be recalled that in oxygen the volatility of radium-*C* occurs at a very much higher temperature than in hydrogen.<sup>37</sup> The formula of Arrhenius for the variation of velocity of solution of metals in acids with temperature has been tested for the radium active deposit, and has been found to hold approximately. A similar series of experiments with the active deposit of thorium showed that thorium-*B* is dissolved more quickly than thorium-*C* by water, salt solutions, alkalis and acids, but less quickly by organic solvents.<sup>38</sup>

### *Radium.*

*Atomic Weight.*—The value found for the atomic weight of radium by analysis of the chloride,<sup>39</sup> 225·95, was confirmed by

<sup>35</sup> T. Godlewski, *Le Radium*, 1913, **10**, 250; *A.*, ii, 1011.

<sup>36</sup> E. Ramstedt, *ibid.*, 159; *A.*, ii, 659.

<sup>37</sup> *Ann. Report*, 1912, 318.

<sup>38</sup> C. F. Hogley, *Phil. Mag.*, 1913, [vi], **25**, 330; *A.*, ii, 273.

<sup>39</sup> Compare *Ann. Report*, 1912, 289.

converting the material into the bromide, and fractionally crystallising it numerous times. The value found in one set of determinations from the ratio  $\text{RaBr}_2/\text{AgBr}$  was 225.96, and in another, from the ratio  $\text{RaBr}_2/\text{Ag}$ , 225.97. A spectroscopic examination of the final product showed no trace of barium. In the mother liquors, barium was detected, and its quantity estimated as 0.03 per cent. From this it is calculated that in the preparations used the percentage of barium in the chloride could not exceed 0.004, and in the bromide 0.002, which is insufficient to affect the accuracy of the atomic-weight determinations. A similar degree of purity may be assumed for the International Standard, so that, before the barium can cause any error, the radioactive methods of comparing standards must be increased in accuracy a hundredfold.<sup>40</sup> The atomic weight being so near the whole number suggests that the atomic weight of uranium should be very nearly 238 instead of 238.5, as in the international list, if mass is conserved in radioactive changes.

*Valency.*—The bivalency of radium has been shown by a new method, depending on the volume of liquid transported in a capillary tube by electric endosmose, under the application of considerable electric potentials. The interest of the method consists in the excessively minute amounts, 0.01 to 0.001 milligram of substance required.<sup>41</sup>

*Estimation.*—A method for the rapid estimation of radium by the emanation method depends on the use of a micro-furnace, in which a tiny fragment of the mineral is heated for a few seconds to 2000° or 3000° in a cavity in a thin carbon rod, inside a small chamber provided with a rubber balloon to allow for expansion. Rapid escape of emanation commences at 750°, and is complete at bright redness.<sup>42</sup>

*Technical Extraction.*—The reduction of the radiferous “raw sulphates” has been effected by mixing the absolutely dry, finely powdered material with finely powdered calcium hydride, and firing the mass with magnesium ribbon and a priming mixture, as in the thermite reaction. The reduction is complete in a few minutes, and the product is leached out with acid as quickly as possible to avoid oxidation of the sulphides with air.<sup>43</sup> It may be doubted whether such a reaction could be technically advantageous on account of the high cost of the reducing agent.

<sup>40</sup> O. Hönigschmid, *Monatsh.*, 1913, **34**, 283; E. Haschek and O. Hönigschmid, *ibid.*, 351; *A.*, ii, 268; S. Meyer, *Physikal. Zeitsch.*, 1913, **14**, 124; *A.*, ii, 267.

<sup>41</sup> H. Friedlich and G. von Elissafoff, *Physikal. Zeitsch.*, 1913, **14**, 1052; *A.*, ii, 1008.

<sup>42</sup> A. L. Fletcher, *Phil. Mag.*, 1913, [vi], **26**, 674; *A.*, ii, 904.

<sup>43</sup> E. Ebler and W. Bender, *Zeitsch. anorg. Chem.*, 1913, **83**, 149; *A.*, ii, 904.

*Uranium-Radium Ratio.*—A new determination of the ratio of radium to uranium in a variety of minerals, in which the uranium was estimated chemically and the radium by both the emanation and  $\gamma$ -ray methods, by the use of standards checked against the international standard, gives the value  $3.328 \times 10^{-7}$ .<sup>44</sup> A review of all the evidence available by which the periods of uranium and radium may be calculated has shown that the different methods lead to substantial agreement, and that the half-periods, respectively, are  $5 \times 10^9$  and 1730 years, corresponding with the average-life periods,  $7.25 \times 10^9$  and 2500 years.<sup>45</sup> The half-periods for thorium and uranium, calculated from the range of the  $\alpha$ -particle and the ionisation per unit weight, have been independently found to be  $1.8 \times 10^{10}$  and  $5 \times 10^9$  years.<sup>46</sup>

*Various.*—The effect of very low temperatures on the penetrating radiation from radium has been carefully studied. Immersion for 1.5 hours in liquid hydrogen did not produce any variation attaining 0.1 per cent., and probably not 0.02 per cent.<sup>47</sup> By an ingenious application of the principle of the radium-clock, in which the negative charge, carried by the penetrating rays, is used to charge a tube containing radium positively, very high potentials have been obtained. A tube of radium emanation was suspended by an insulating support in a high vacuum, and the potential it attained measured by a simple form of attracted disk electrometer. The tube quickly charged itself to a high potential. In spite of all precautions to obtain as perfect a vacuum as possible, the attainable potential was limited to a maximum of about 150,000 volts, at which a discharge always occurred.<sup>48</sup>

#### *The Actinium Series.*

*Period of Actinium-X.*—A small change has to be recorded in the period of actinium-*X*, which still, however, remains an exception to the Geiger-Nuttall relation, the earlier reported resolution of radioactinium into two products having been withdrawn.<sup>49</sup> From  $\beta$ -ray measurements, the half-period of actinium-*X*, very carefully purified, was found to be 11.6 days, corresponding with the average life period, 16.8 days. For radioactinium the accepted values were

<sup>44</sup> B. Heimann and W. Marcwald, *Physikal. Zeitsch.*, 1913, **14**, 303; *Jahrb. Radioakt. Elektronik*, 1913, **10**, 299; *A.*, ii, 374.

<sup>45</sup> S. Meyer, *Sitzungsber. K. Akad. Wiss. Wien*, 1913, **122**, (iia), 1087.

<sup>46</sup> H. N. McCoy, *Physical Review*, 1913, **1**, 393.

<sup>47</sup> Mme. M. Curie and H. K. Onnes, *Le Radium*, 1913, **10**, 181; *A.*, ii, 746.

<sup>48</sup> H. G. J. Moseley, *Proc. Roy. Soc.*, 1913, **A**, **88**, 471.

<sup>49</sup> *Ann. Report*, 1912, 294; O. Hahn and L. Meitner, *Physikal. Zeitsch.*, 1913, **14**, 752; *A.*, ii, 821; A. S. Russell and J. Chadwick, *Phil. Mag.*, 1914, [vi], **27**, 112.

confirmed (19.5 and 28.2 days).<sup>50</sup> No  $\beta$ -radiation could be detected from the very carefully purified actinium. In another determination, by  $\alpha$ -ray measurements, the value 11.35 days for the half-period of actinium-*X* was obtained.<sup>51</sup>

### *$\alpha$ -Rays.*

Experiments on the scattering of the  $\alpha$ -rays by gases<sup>52</sup> and metal foils<sup>53</sup> have been continued, with results in agreement with the requirements of Rutherford's theory of the structure of the atom (p. 270). The theory has been quantitatively tested, as regards the variation of the number of  $\alpha$ -particles deflected when (1) the angle of scattering, (2) the thickness of the foil, (3) the atomic weight of the scattering material, and (4) the velocity of the  $\alpha$ -rays are varied. Although the results are approximately in agreement with the view that the positive nuclear charge is one-half of the atomic weight, it is probable that they would be more perfectly in agreement with van den Broek's view that the charge is equal to the atomic number, which for heavy elements is somewhat less than half the atomic weight.

Attention may be directed to a comparison of the ranges of the  $\alpha$ -rays of radium-*C* and polonium in hydrogen and helium respectively,<sup>54</sup> since this is, theoretically, a very interesting case of a denser gas allowing the  $\alpha$ -particles to travel in it slightly further than in a lighter gas. The ranges of the  $\alpha$ -ray of radium-*C* in the two gases at N.T.P. are, respectively, 29.36 cm. in hydrogen, and 30.84 cm. in helium. This furnishes the simplest possible proof, as previously pointed out, of the monatomicity of the helium molecule. For, on Bragg's law, the atomic stopping power is approximately proportional to the square root of the atomic weight. If the helium molecule is monatomic, the number of atoms in the path of the  $\alpha$ -ray will be twice as great in hydrogen as in helium, and the stopping power of each atom twice as great in helium as in hydrogen, whereas any other assumption must make the range in helium less than in hydrogen.

*Pleochroic Haloes.*—An attempt has been made to arrive at the age of the pleochroic haloes occurring in the mica of Haughtonite, co. Carlow, which contains no thorium haloes, but numerous uranium haloes of all stages of development, from the smallest, due

<sup>50</sup> O. Hahn and M. Rothenbach, *Physikal. Zeitsch.*, 1913, **14**, 409; *A.*, ii, 463.

<sup>51</sup> H. N. McCoy and E. D. Leman, *ibid.*, 1280; *A.*, 1914, ii, 17.

<sup>52</sup> E. Rutherford and J. M. Nuttall, *Phil. Mag.*, 1913, [vi], **26**, 702; *A.*, ii, 898.

<sup>53</sup> H. Geiger and E. Marsden, *ibid.*, 1913, [vi], **25**, 604; *A.*, ii, 371.

<sup>54</sup> T. S. Taylor, *ibid.*, 1913, [vi], **26**, 402; *A.*, ii, 899.

to the rays of uranium and ionium only, to the largest due to the fully darkened haloes, completed to the range of the  $\alpha$ -rays of radium-*C*.<sup>55</sup> The number of  $\alpha$ -particles required to produce a certain density of staining in the mica was experimentally determined, the mass of the nucleus giving the halo was estimated microscopically from its dimensions, and the quantity of radium per unit of mass assumed from analogy to the radium content of large zircons and cyrtolite. The last is, of course, the uncertain factor, and the results were dealt with in all cases to yield the lower limit possible for the age. The results are interesting in that they confirm the somewhat excessive periods of age given by the helium and lead contents of uranium minerals. The haloes may most safely be regarded as of early Devonian age, and the results point to a period of not less than four hundred million years as being required in their formation.

#### *$\beta$ -Rays.*

The analysis of the slower  $\beta$ -rays given by radium-*B* and radium-*D* showed in each case four distinct beams of velocity, in terms of that of light, 0.361, 0.362, 0.412, 0.422 for the first, and 0.342, 0.344, 0.390, and 0.402, for the second substance. The differences between the energies of the several groups of rays are the same for each substance, within the error of measurement. This leads to the conclusion that in each case the rays are produced by the same mechanism.<sup>56</sup> Taken in combination with the isotopism of the two substances, the result is very suggestive. An exhaustive analysis has been made of the very complex  $\beta$ -radiations of radium-*B* + *C*, from photographs taken by a new method, which enables a comparatively wide slit to be employed without loss of sharpness.<sup>57</sup> The velocity and energy for 16 lines from radium-*B* and 48 lines from radium-*C* have been tabulated.

With regard to radium-*B*, no lines were observed with a velocity above 0.832, although it is probable that faster beams exist which are masked by the stronger lines of radium-*C*. The slowest line observed had a velocity of 0.365. Two complex groups of low velocity lines shown strongly by radium-*B* were shown faintly by radium-*C*. For radium-*C* 29 lines were observed over the range of velocity from 0.9858 to 0.946, the energy of which were all integral multiples of  $E (= 0.428 \times 10^{13}e)$ . The integers were the alternate odd integers from 59 to 47, and, with the exception of 45, every

<sup>55</sup> *Ann. Report*, 1910, 260; J. Joly and E. Rutherford, *Phil. Mag.*, 1913, [vi], 25, 644.

<sup>56</sup> J. Danysz, *Le Radium*, 1913, 10, 4; *A.*, ii, 270.

<sup>57</sup> E. Rutherford and H. Robinson, *Phil. Mag.*, 1913, [vi], 26, 717; *A.*, ii, 899.

integer between 46 and 24. Below this, the differences of energy between the lines were much smaller, and the plates were crowded with fine lines extending right into the low velocity regions where radium-*B* shows strong lines. No simple relation appears to hold between the energies of the radium-*B* lines. This is probably connected with the fact that whereas the  $\gamma$ -radiation from radium-*C* is homogeneous, that from radium-*B* consists of three widely-different types. The quantity  $E$  represents, on Rutherford's theory of the origin of  $\gamma$ -rays, the energy abstracted from the  $\beta$ -particle in passing through certain regions of the atom and converted into  $\gamma$ -ray energy.

There is one point of interest in connexion with the expulsion of  $\alpha$ -rays. In the cases most closely studied the recoil product also carries a single positive charge,<sup>58</sup> so that three positive charges result. Two of these are accounted for by internal compensation within the atom, the outward sign of which is the shift of two places in the periodic table. The third is not yet accounted for. An attempt to detect the expulsion of a soft  $\beta$ -ray from radium-*A* led to no result.<sup>59</sup> It is quite possible, however, that the third positive charge may be compensated for by the loss of a  $\delta$ -ray, which would not be detectable in ordinary circumstances. Radium,<sup>60</sup> thorium-*X*, and radioactinium do give soft  $\beta$ -rays in addition to  $\alpha$ -rays, and only the latter are accounted for in the change of place in the periodic table. On the other hand, the two rayless changes, of mesothorium-*I* and actinium, are equivalent to  $\beta$ -ray changes in the alteration of place they produce. These points, which are of the nature of exceptions to the simple generalisation, call for fuller examination.

### *$\gamma$ -Rays.*

The chief centre of interest in the physical examination of the various types of rays has been successively, first the  $\alpha$ -rays, then the  $\beta$ -rays, and now the  $\gamma$ -rays. On the one hand, the view has triumphed that the  $\gamma$ -radiation is essentially homogeneous. This view was put forward after the exhaustive examination of the  $\gamma$ -rays of radium in various materials, from a thickness equivalent to 1 cm. of lead up to 22 cm. A repetition with larger quantities of radium, in which mercury was used as the absorbent, showed a strictly exponential absorption between the thickness of from 1 to 22.5 cm. The value found for  $\mu/d$ , the absorption-coefficient

<sup>58</sup> *Ann. Report*, 1910, 272; H. P. Walmsley, *Phil. Mag.*, 1913, [vi], 26, 401; *A.*, ii, 905.

<sup>59</sup> W. Makower and S. Russ, *Proc. London Phys. Soc.*, 1913, 25, 253; *A.*, ii, 654.

<sup>60</sup> Compare L. Kolowrat, *Le Radium*, 1913, 10, 280.

divided by the density, was  $4.38 \times 10^{-2}(\text{cm.})^{-1}$ , in almost absolute agreement with the former value found for lead.<sup>61</sup> No evidence of an ionising radiation more penetrating than the known  $\gamma$ -rays was obtained. Through 30 cm. of mercury it can be calculated that no radiation could be detected, even from 10 grams of radium, so that it would be thought that the equivalent of the 76 cm. which surrounds us as an atmosphere must screen us absolutely from any cosmical source of  $\gamma$ -rays that exists (p. 288).

On the other hand, the departure of the absorption curves from the simple exponential type, for small thicknesses of material, has been shown to be due to the existence of a large number of types of homogeneous  $\gamma$ -radiation, varying in penetrating power from those of thorium-*D*, for which  $\mu/d$  is  $3.5 \times 10^{-2}(\text{cm.})^{-1}$ , to one of the types given by radium-*B*, for which  $\mu/d$  is 85, which are hardly more penetrating than  $\alpha$ -rays.<sup>62</sup> So that, just as in the case of the  $\beta$ -rays, fuller examination has shown that the most obvious more penetrating types are by no means the only ones that exist. Twenty types have so far been distinguished, as set forth with the values of  $\mu/d$  in aluminium for each type, in the following table:

| Element.                  | At. wt. | $\mu/d$ (Al).                                       | Element.                 | At. wt. | $\mu/d$ (Al).               |
|---------------------------|---------|---|--------------------------|---------|-----------------------------|
| Radium- <i>B</i> .....    | 214     | 85.0 }<br>14.7 }<br>0.188 }                         | Thorium- <i>B</i> .....  | 212     | 59.0 }<br>11.8 }<br>0.13 }  |
| Radium- <i>C</i> .....    | 214     | 0.0424  | Thorium- <i>D</i> .....  | 208     | 0.035                       |
| Radium- <i>D</i> .....    | 210     | 16.5 }<br>0.36 }                                    | Radio-actinium ...       | —       | 9.2 }<br>0.070 }            |
| Radium- <i>E</i> .....    | 210     | similar to<br>radium- <i>D</i> , but<br>very feeble | Actinium- <i>B</i> ..... | —       | 44.0 }<br>11.4 }<br>0.165 } |
| Mesothorium- <i>II</i> .. | 223     | 9.5 }<br>0.043 }                                    | Actinium- <i>D</i> ..... | —       | 0.073                       |

Four types may be distinguished in descending order of penetrating power: (1) The known, or old-fashioned, most penetrating type, with  $\mu/d$  varying from 0.035 to 0.073. The elements long known as  $\gamma$ -ray producers, radium-*C*, thorium-*D*, and actinium-*D* are unique in giving only this single homogeneous type. In addition mesothorium-*II* and radioactinium give a  $\gamma$ -ray of this type, in each case accompanied by another of type (3). (2) Next in penetrating power are some fairly penetrating rays, for which  $\mu/d$  varies from 0.13 to 0.188 given by the *B*-members only and (4) feeblest in penetrating power of all another set,  $\mu/d$  44 to 85, also given only by the *B*-members. There remains to consider (3) a type, with  $\mu/d$  varying from 9.2 to 16.5, given by all the elements which

<sup>61</sup> A. S. Russell, *Proc. Roy. Soc.*, 1913, A, 88, 75; *A.*, ii, 270; compare also A. Brömmer, *Sitzungsber. K. Akad. Wiss. Wien.* 1912, 121, [ia], 1563.

<sup>62</sup> E. Rutherford and H. Richardson, *Phil. Mag.*, 1913, [vi], 25, 722; 26, 324, 937; *A.*, 1913, ii, 461, 901; 1914, ii, 13.

give  $\gamma$ -rays at all, except radium-*C*, thorium-*D*, and actinium-*D*. This type is probably the characteristic *X*-radiation of the respective elements of the *L*-series, type (1) being probably also characteristic *X*-rays of the *K*-series. Type (3) are all less penetrating than hard  $\beta$ -rays and than the *X*-rays of an average tube. Type (4) are of the same order of penetrating power as low-velocity  $\beta$ -rays, and are not greatly superior in this respect to  $\alpha$ -rays.

Thorium-*X* also gives a soft  $\gamma$ -radiation, intermediate in penetrating power between the  $\beta$ - and  $\gamma$ -rays of thorium-*D*.<sup>63</sup> If thorium-*C*, which gives the greater and more penetrating part of the  $\beta$ -rays of thorium-*B* + -*C*, gives any  $\gamma$ -radiation at all, it must be of exactly the same penetrating power as that of thorium-*D*. Radium, which, like thorium-*X*, is anomalous in giving some soft  $\beta$ -rays as well as  $\alpha$ -rays, has now been found to emit  $\gamma$ -rays also, of intensity about 1 to 1.5 per cent. of that emitted by radium in equilibrium with radium-*B* + -*C*.<sup>64</sup> The radiation appears to consist of three types with  $\mu/d$  130, 6, and 0.1.

Further details have been published of the  $\gamma$ -rays excited by the  $\alpha$ -rays in elements of high atomic weight,<sup>65</sup> and evidence of such radiation obtained for radiothorium as well as from ionium. Polonium emits a single type of  $\gamma$ -radiation ( $\mu/d=215$ ) due to the polonium itself. When deposited on copper, but not on aluminium, lead, or platinum, a second  $\gamma$ -radiation is present, which is probably the characteristic radiation of copper, in the *L*-series. A very small quantity of  $\beta$ -rays is also emitted from polonium, which could not be ascribed to impurity.

No doubt, when all the details are worked out for the  $\beta$ - and  $\gamma$ -rays, much new light will be thrown on the structure of the atom and the mechanism at work in radioactive processes. A beginning has already been made in the method of studying  $\gamma$ -rays by reflection from crystal surfaces. The  $\gamma$ -rays from radium-*B* ( $\mu/d=14.7$ ) are reflected from a rock salt crystal at angles between  $8^\circ$  and  $10^\circ$ , as a group of fine lines, whereas another series of fine lines were observed reflected at an angle of about  $2^\circ$ , probably resulting from the more penetrating rays of radium-*B* and -*C*.<sup>66</sup>

<sup>63</sup> L. Meitner and O. Hahn, *Physikal. Zeitsch.*, 1913, **14**, 873; *A.*, ii, 906.

<sup>64</sup> A. S. Russell and J. Chadwick, *Phil. Mag.*, 1914, [vi], **27**, 112.

<sup>65</sup> *Ann. Report*, 1912, 300; J. Chadwick, *ibid.*, 1913, [vi], **25**, 193; *A.*, ii, 91; J. Chadwick and A. S. Russell, *Proc. Roy. Soc.*, 1913, A, **88**, 217; *A.*, ii, 372; *Phil. Mag.*, 1914, [vi], **27**, 112.

<sup>66</sup> E. Rutherford and E. N. da C. Andrade, *Nature*, 1913, **92**, 267.

*Transmutational Reactions.*

There is nothing new to report concerning the question of transmutational reactions produced by the radium emanation.<sup>67</sup> On the other hand, the possibility of a synthesis of helium and neon under the action of the electric discharge has been much discussed. Helium has been obtained from old X-ray bulbs, which, after prolonged use, were broken up and heated to redness.<sup>68</sup> The spectrum of the evolved gases, after treatment with cold charcoal, showed the helium lines with great brilliance, and some of the brighter neon lines faintly. In another experiment unopened X-ray bulbs were connected by rubber tube to a pump, and, after exhaustion of the connecting tube, communication between the bulb and pump was established by breaking the seal under the rubber. The bulb was heated to 350° and washed out with oxygen, which again showed the presence of helium, and a just detectable trace of neon. On then breaking up these bulbs and heating them, no trace of gas, not absorbed by cold charcoal, was obtained. This makes it appear probable that the helium is in some way produced by the discharge in the tubes, and only loosely held by the glass in a form readily and completely removable by heat.

The work on the production of neon by bombarding fluorite by cathode-rays, referred to briefly last year,<sup>69</sup> has now been published in detail.<sup>70</sup> It was found that neon was obtained, even in absence of fluorite, when the discharge passed in hydrogen. The hydrogen was exploded with excess of oxygen, and treated with cold charcoal. In one experiment the amount of neon was of the order of that obtained in 1 or 2 c.c. of air—that is, of the order of 0.01 cu.mm.—but no nitrogen was found in the gas absorbed by the charcoal. Oxygen and hydrogen put through the pump in the same way as during an experiment, without being submitted to the action of the discharge, gave no neon, neither could it be obtained from the glass and electrodes by heating. From an exhausted bulb, heated in the air to the softening point, no neon was obtained, and to test whether glass subjected to the cathode discharge might be porous to neon, a bulb was completely surrounded by another attached to a separate pump. The cathode-ray discharge was passed through hydrogen in the inner bulb, with the outer bulb exhausted, for two

<sup>67</sup> *Ann. Report*, 1909, 255.

<sup>68</sup> Sir William Ramsay, *T.*, 1913, **103**, 264.

<sup>69</sup> *Ann. Report*, 1912, 308. The reference there given to Sir William Ramsay's letter to *Nature*, in which this result is quoted, is misleading in that it omitted to mention that the author of the work was Prof. Collie, who had not, at the time, published the result himself.

<sup>70</sup> J. N. Collie and H. S. Patterson, *T.*, 1913, **103**, 419.

days, and the usual amount of neon was found in the hydrogen. Now, however, the surprising observation was made that when the outer bulb was washed out with oxygen, a faint explosion on sparking showed the presence of hydrogen, and the residue, after cold charcoal absorption, was about fifty times greater than the residue from the inner tube. This gas was mainly helium with enough neon present to give the spectrum. At this stage the author was joined by H. S. Patterson, who had obtained similar quantities of neon independently, by very similar methods. Pure hydrogen sparked in a Plücker tube and pumped out whilst the discharge was continued showed, after explosion with oxygen and absorption with cold charcoal, the presence of neon, sometimes containing helium, but always with the helium in much greater relative quantity than in the helium-neon mixture obtained by cold charcoal from air. The jacketed tube experiments were also repeated with the same results, and it was found that, when the outer tube contained oxygen, almost pure neon, instead of almost pure helium, was obtained from the gas in the outer vessel.

In continuation of the work,<sup>71</sup> the joint investigators found that electrodes were not necessary for the production of neon, which was formed in hydrogen, free from mercury vapour, submitted to the electrodeless discharge. The jacketed tube experiments were also repeated with the wires connecting the electrodes of the inner tube encased in glass tubes. The apparent disappearance of 3.6 c.c. of hydrogen under the action of the discharge was observed. The gas from the outer tube, examined by filling the latter with mercury and compressing the gas into a spectrum tube, gave a "carbon spectrum." On sparking, it decreased in volume, and the carbon spectrum disappeared. Even after absorption in cold charcoal, which should leave pure helium and neon, this carbon spectrum is seen. The gas causing it appears to be highly uncondensable and not readily oxidised, and it is suggested it may be the " $X_3$ " discovered by Sir J. J. Thomson by his positive ray method. Another observer, with the jacketed tube experiment, found only hydrogen, containing small and varying amounts of carbon monoxide, accumulating in the jacket after discharge in the inner tube. The production of neon from hydrogen sparked in the inner tube ceased after a time, but, on adding a mixture of hydrogen and oxygen and continuing the discharge, the production of neon recommenced.<sup>72</sup> These results have been given fully because it is difficult at the present stage to compress or comment on them. That neon is not always obtained by sparking hydrogen is shown by recent experi-

<sup>71</sup> J. N. Collie and H. S. Patterson, *P.*, 1913, 29, 217.

<sup>72</sup> J. I. O. Masson, *ibid.*, 233.

ments, of which only the abstract is available at the time of writing.<sup>73</sup> This is in accord with experience of the writer with the calcium method, in which the production of neon from hydrogen under the electric discharge could not fail to have been observed if it were a general phenomenon. There must be some unrecognised factor contributing to the results. In view of the care and circumspection apparently exercised in the performance of the experiments, it would not be fair to conclude that the neon found must necessarily have been of atmospheric origin.

Helium and neon have been commonly found in the positive-ray experiments,<sup>74</sup> but here, where apparently a continuous stream of gas is kept passing through the discharge tube in connexion with a cold charcoal bulb, it must be very difficult to be sure that they are not derived from the atmosphere. In these experiments it was found that the conditions which gave rise to a considerable amount of "X<sub>3</sub>" generally gave helium and neon also. The view now most favoured with regard to the nature of "X<sub>3</sub>" is that it is probably a modification of hydrogen (H<sub>3</sub>). It is obtained by the cathode-ray bombardment of many substances, indeed, most substances, but especially abundantly from potassium salts. The production of hydrogen under the discharge in vacuum tubes is regarded as analogous to the production of "X<sub>3</sub>," helium and neon, and the suggestion is made that they may represent abortive attempts at disintegration, analogous to those occurring in radioactive substances, in which the products require the assistance of the cathode-ray bombardment in order to get clear from the atoms producing them.

#### *Chemical and Biochemical Effects of Radioactivity.*

A very thorough investigation of the decomposition of water by  $\alpha$ -rays has been made. For ice at  $-183^{\circ}$  the products are hydrogen and oxygen in the same proportion as in water, but the amount of decomposition is only a twentieth as great as with liquid water. For the latter, 6.4 per cent. of the energy available is utilised in the decomposition, and the number of molecules of water decomposed is 6 per cent. greater than the number of ions produced by the rays in air. At first hydrogen is produced in excess with formation of hydrogen peroxide. Later there is a disengagement of excess of oxygen. When the rays act on water in the state of vapour, the excess of hydrogen attains 50 per cent. by volume.<sup>75</sup>

These results therefore form an important confirmation of the

<sup>73</sup> Hon. R. J. Strutt, *Proc. Roy. Soc.*, 1914, *A.*, **89**, 499.

<sup>74</sup> Sir J. J. Thomson, *Nature*, 1913, **90**, 645.

<sup>75</sup> W. Duane and O. Schuer, *Le Radium*, 1913, **10**, 33; *A.*, ii, 270.

view that the number of molecules of liquid water decomposed by the passage of an  $\alpha$ -ray is approximately the same as the number of pairs of ions produced by the  $\alpha$ -ray in passage through a gas; or, in other words, if the ionisation current in the gas were expressed in faradays, the decomposition in liquid water would be expressed in moles.

Researches on the decomposition of iodides by the penetrating radium rays have been extended to the iodides of the alkaline-earth elements, which show the same general behaviour as those of the alkali metals.<sup>76</sup> Although the effects produced are of a smaller order of magnitude than those observed with ultra-violet light, they are of the same general character. Thus, with ultra-violet light, many organic acids in  $N/2$ - to  $2N$ -solution were decomposed, acetic acid to the greatest extent, but with long exposures to the penetrating rays of comparatively large quantities of radium, the effects are excessively minute. Among positive actions of the latter may be mentioned the reduction of silver nitrate solution to metallic silver, the oxidation of ethyl alcohol to acetaldehyde and acetic acid, and the inversion of sterilised aqueous solutions of sucrose.<sup>77</sup> A further study has been made of the action of radium emanation on growing plants.<sup>78</sup> A lasting injury is caused to the plant, whether the exposure is made to the seed or to the seedling, provided that the emanation is of sufficient concentration, and this injury, unlike that caused by tobacco-smoke or coal-gas, continues after the plant has been brought into pure air. In smaller quantities the emanation exerts in some cases a favourable influence. The injury extends not only to the growing parts, but also to the organs already developed, including the leaves. The fall of the leaves is, in certain cases, much accelerated by the emanation, and can be made to occur in spring or summer, when under normal conditions no such tendency exists. Lastly, remarkable changes sometimes occur in the character of the development. The buds of *Sedum Sieboldii*, instead of developing normally into trifoliated whorls, after exposure, in a quite early stage of growth, to strong emanation for three days, develop with decussate pairs of leaves. In some ways the action of the emanation resembles that of a poison, but there is probably no poison known capable of producing such great effects in such infinitesimal quantity.

<sup>76</sup> *Ann. Report*, 1912, 323.

<sup>77</sup> A. Kailan, *Monatsh.*, 1913, **34**, 359, 1209, 1245, 1269; *A.*, ii, 270, 1000, 1002, 1003.

<sup>78</sup> H. Molisch, *Sitzungsber. K. Akad. Wiss. Wien*, 1912, **121**, [iia], 833.

*The Penetrating Radiation of the Atmosphere.*

In seven balloon ascents in 1912 measurements of the ionisation in a closed electroscope were carried out, and the results show that there is a very small decrease of the penetrating radiation between 200 and 1000 metres, and then an increase, so that at heights from 1000 to 2000 metres the radiation has much the same value as at the surface. In one ascent, in which a height of 5350 metres was attained, a very considerable increase of the radiation was observed above 3000 metres, and the increase became very marked at the greatest height attained. This increase above 3000 metres was independently confirmed, and in 1913 in another voyage, in which 4160 metres was attained, the same slow increase from 2000 to 3500 metres and rapid increase at the highest level was observed, although the weather conditions were quite different from that of the earlier ascents. The conclusion is drawn from this that a very penetrating radiation must enter the upper atmosphere from outside space, and cause part of the spontaneous ionisation in closed vessels observed at the surface. This part is probably that for which large and irregular variations have been noticed, but as no diminution was observed in the ascents at night and during a solar eclipse, the source of this radiation can scarcely be the sun. It is estimated that only one-twentieth part of the penetrating radiation observed at 1000 to 2000 metres height can be due to the radium-*C* in the atmosphere, assuming that this is no greater than at the surface. At this height the rays from the earth's surface would be so much absorbed by the air as to be negligible. The conclusion is a somewhat sensational one. When it is considered that the atmosphere is equivalent to 76 cm. of mercury, and would reduce the  $\gamma$ -radiation of radium to between  $10^{-19}$  and  $10^{-20}$  of the initial value, the assumption of an external radiation reaching the surface of the earth from outer space would seem to involve the existence either of more penetrating rays than are yet known, or of cosmical sources of radioactivity of unparalleled intensity.<sup>79</sup>

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<sup>79</sup> V. F. Hess, *Sitzungsber. K. Akad. Wiss. Wien*, 1912, **121**, [iia], 2001; 1913, **122**, [iia], 1053, 1481.