RADIOACTIVITY.¹

α -Rays.

IN previous Reports the evidence has been given for the conclusion that the α -radiation from radioactive substances is due to the expulsion of positively charged particles of atomic dimensions with a velocity of the order of about one-fifteenth that of light. The initial velocity of expulsion is the same for all the α -particles of any one type of radioactive matter, but differs, within somewhat narrow limits, for the different types. The mass, however, is identical in all cases, and is $2n$ times that of hydrogen, where n is the number of atomic charges carried by the α -particle, which may be compared with the valency of an ion in electrolysis. Methods have now been developed which have enabled the number of α -particles expelled from a radioactive substance to be directly counted, and these have established that $n=2$. The atomic weight, therefore, of the α -particle is 4. Other evidence has proved directly the identity of the α -particle with the helium atom, a result long indirectly foreshadowed. The theory of probability applied to radioactive changes² leads to the conclusion that, when the numbers of atoms disintegrating in unit time is few, variations from the average number may be anticipated, inversely proportional in magnitude to the square root of that number. Such rapid variations in the intensity of α -radiation, and therefore of the ionisation, from a constant source, in part observed unwittingly,³ in part sought for deliberately⁴ make themselves apparent, especially when the ionisation currents of two separate similar preparations are balanced against each other. Complete balance is never so attained, the indicating instrument showing perpetual excursions on either side of the zero position. This phenomenon has now been thoroughly studied,⁵ and found to be due to real variations in the rate of

¹ This report covers the years 1908 and 1909.

² E. von Schweidler, *Beibl. Ann. Physik.*, 1907, **31**, 356.

³ H. L. Bronson, *Phil. Mag.*, 1906, [vi], **11**, 143.

⁴ K. W. F. Kohlrausch, *Sitzungsber. K. Akad. Wiss. Wien*, 1906, **115**, (iia), 673.

⁵ E. Meyer and E. Regener, *Ann. Physik*, 1903, [iv], **25**, 757; H. Geiger, *Phil. Mag.*, 1908, [vi], **15**, 539.

expulsion of α -particles, which have been shown, although the result has been subject to some criticism,⁶ to follow the law deduced from the theory of probability.

Two independent methods have been now developed which are sufficiently sensitive to enable the effect of a single α -particle to be recorded. The first⁷ makes use of the scintillations produced in zinc sulphide, the discontinuity of which in the spinthariscopes has long been known. A definite small portion of the zinc sulphide screen, faintly illuminated to assist in focussing, is viewed through a microscope of suitable aperture and magnification, so that the picture appears in not less than its natural brightness. Under these conditions, for which the original paper may be profitably consulted, the number of scintillations in the field of vision can readily be counted, when the intensity of the source is such that they do not exceed more than one every few seconds. In the second method,⁸ which is less simple, but offers more guarantee that all the α -particles expelled are counted, the ionisation in the gas produced by the α -particle is magnified by the application of a potential at the electrodes so great that the gas is near the breaking-down point at which a disruptive discharge would pass. In these circumstances the ions produced themselves become ionising agencies—the negative ions more readily than the positive—by virtue of the kinetic energy acquired under the field, and produce fresh ions in their path by collisions. The field is adjusted so that only the negative ions so ionise, and the magnification of the effect is then large but not infinite, the energy of the α -particles, as it were, merely pulling the trigger whereby a much larger quantity of the energy of the electric field is released. Fields of the order of 1000 volts per cm., and a gas pressure of about 5 cm., were employed. The α -rays from a powerful source traverse an exhausted tube several metres in length, and pass through a minute aperture along the axis of the detecting chamber, which consists of a narrow cylinder, connected to the high voltage battery, carrying a central electrode connected with the electrometer. The arrival of each α -particle causes the electrometer needle to move with a sudden jerk, after which, by means of a suitable “air resistance,” it automatically returns again to zero. The number of jerks—usually adjusted to one every few seconds—gives the number of α -particles entering the detecting chamber. From geometrical considerations, the total number expelled from the source can thus be deduced.

⁶ N. Campbell, *Proc. Camb. Phil. Soc.*, 1909, **15**, 117.

⁷ E. Regener, *Ber. Deut. physikal. Ges.*, 1908, 78.

⁸ E. Rutherford and H. Geiger, *Proc. Roy. Soc.*, 1908, **81**, A, 141; *A.*, 1908, ii, 555.

In this way, it was found that 3.4×10^{10} α -particles are expelled per second from the radium-*C* in equilibrium with 1 gram of radium. This fundamental constant will be termed *Q*. Each of the first four α -ray changes of radium give the same number of α -particles.⁹ Hence this number holds equally for radium itself, the emanation, and radium-*A*, the total number per second from 1 gram of radium in equilibrium¹⁰ being therefore 1.36×10^{10} per second. Taking the limit of detection of minute quantities of radium by the emanation test as 10^{-12} gram,¹¹ it follows that this corresponds with the expulsion of only one α -particle every 100 seconds, and, indeed, the von Schweidler variation is easily detectable by the irregular motion of the gold-leaf in such measurements.

A comparison of the results of the electric and scintillation methods showed that both gave the same result, so that under suitable conditions each α -particle produces a scintillation. This being the case, it is probable that the scintillation method, on account of its simplicity, will be widely employed, and will supplant the electric method almost completely.

The electric and fluorescence methods have, since the above account was written, been supplemented by a third, depending on the photographic action of the α -rays. It has been shown that each halide grain in the film struck by an α -particle is rendered capable of development, and the method appears to consist in counting the number of opaque grains in the film by means of a microscope. It is claimed that it is applicable to very weak sources by the use of very long exposures, and to α -particles of very short range.¹²

From the total positive charge carried by the α -particles from a known mass of radium, the individual charge on each α -particle was deduced to be 9.3×10^{-10} E.S.U.¹³ This is between two and three times the previously accepted value of the single atomic charge, 3.4×10^{-10} , but there is now good reason for believing that the older determinations, by the well-known cloud method, gave too low a value, and that 4.65×10^{-10} , or one-half of the charge on the α -particle, is nearer the truth. More recent determinations by the cloud method¹⁴ have given 4.65, whilst the study of the Brownian movement in liquids¹⁵ has given 4.1. By the aid of the ultra microscope, the same constant has also been determined for the

⁹ *Ann. Report*, 1905, 296.

¹⁰ Compare *ibid.*, 295 (line 8 from bottom).

¹¹ F. Soddy, *Phil. Mag.*, 1909, [vi], **18**, 851; *A.*, 1910, ii, 10.

¹² S. Kinoshita, Royal Soc. Meeting, 9/12/09 (abstract only available).

¹³ E. Rutherford and H. Geiger, *Proc. Roy. Soc.*, 1908, **81**, A, 162; *A.*, 1908, ii, 794.

¹⁴ Millikan and Begeman, *Physical Rev.*, 1908, 197; R. A. Millikan, *Phil. Mag.*, 1910, [vi], **19**, 209.

¹⁵ J. Perrin, *Ann. Chim. Phys.*, 1909, [viii], **18**, 1.

charged fine dust produced in a gas by the arc between metallic electrodes,¹⁶ and found to be 4.6. From Planck's theory of radiation, the value 4.69 has been found. There is thus a considerable body of evidence¹⁷ that the α -particle carries two atomic charges, and the value of its atomic mass is therefore 4. Dividing the farad by the new value of the atomic charge leads to the result that there are 6.2×10^{23} atoms in one gram of hydrogen. The period of average life of radium, since the atomic weight is known, can be deduced from the number of atoms in one gram and the number of α -particles expelled per second, on the assumption that *only one α -particle* is expelled per atom disintegrating. The value so found (2560 years) agrees so well with the best determinations that it may now be considered definitely proved that only one α -particle is so expelled. The theoretical volume of emanation in equilibrium with one gram of radium, on the assumption that one molecule of mon-atomic emanation results from one atom of radium disintegrating, is 0.585 cu. mm. at N.T.P., whereas the theoretical rate of production of helium, four atoms of helium resulting from each atom of radium disintegrating, is 158 cu. mm. per year. The theoretical heating effect of the α -particles from one gram of radium is 113 gram cal. per hour. These fundamental quantities, calculated from the value of Q , are all so completely borne out by experiment, that no doubt remains of the essential accuracy of the underlying assumptions.

The identity of the α -particle with helium has also been proved in the most complete manner.¹⁸ The emanation from 140 milligrams of radium was stored in exceedingly thin-walled glass tubes surrounded by an outer exhausted jacket. The tubes were proved to be perfectly gas-tight and impervious to the passage of ordinary gaseous helium, but their thickness was less than the range of the α -particle in glass, so that practically all the α -particles expelled from the enclosed emanation penetrated, with diminished velocity, the glass walls of the tube. The presence of helium in the outer surrounding jacket was tested for by filling it with mercury and compressing its contents into a tiny spectrum tube. In this way the passage of helium in the form of α -particles through the gas-tight glass tube was decisively shown. As is to be expected, the α -particles, owing to their velocity of projection, bury themselves to a slight depth in the wall of the outer jacket, and an appreciable time is taken for the helium to diffuse out. By wrapping lead foil round similar thin tubes, filled with emanation, and not surrounded by a jacket but left exposed to the air, and subsequently melting the

¹⁶ F. Ehrenhaft, *Physikal. Zeitsch.*, 1909, 10, 308.

¹⁷ Compare also M. Moulin, *Le Radium*, 1909, 6, 164.

¹⁸ E. Rutherford and T. Royds, *Phil. Mag.*, 1909, [vi], 17, 281; *A.*, ii, 203.

lead in a charcoal vacuum, it was proved that the lead retains some of the helium it receives during its bombardment with α -particles. The same lead not so treated gave no helium when melted. This removes the last possible objection that the helium was in the glass all the time and merely liberated by the action of the α -particles.

In view of the simplicity and directness of the radium series of changes, each successive α -ray disintegration giving one atom of helium from each atom undergoing disintegration, it might be concluded that other α -ray changes would be similar. It is, however, in the highest degree significant that this does not appear to be the case. The relative ionisation given by the emanation and the active deposit of thorium and actinium respectively¹⁹ is in each case such as would result if twice as many α -particles were given off in the change of the emanation as in the change of the active deposit. As there is conclusive indirect evidence²⁰ that in the change of the thorium active deposit two α -particles of different range are emitted, probably in consecutive changes so rapid that they still defy analysis, it appears that four α -particles at least must result from one atom of thorium emanation on disintegration. So far, the α -rays of the active deposit of actinium have been considered homogeneous, and due to actinium-*B*, but anomalies in the α -ray range curves have now been observed²¹ for this case also, and a similar explanation to that for the thorium active deposit has been suggested. Here, however, the difference in the ranges can only amount to a few millimetres of air, and the matter is not so definite. If the explanation is correct, the actinium emanation must also give at least four α -particles. The case of uranium, which appears to give two α -particles, will be again referred to. The number of ions (presumably pairs of ions are referred to) produced by the α -particle of radium-*C* during its whole range in air has been redetermined²² on the new data to be 2.37×10^5 . The specific ionisation at each point of the range has been determined. At the end of the range this diminishes very rapidly, but still not absolutely abruptly. The data obtained allows the number in the case of any other α -particle of different range to be calculated. The estimation of small quantities of radium by means of the saturation ionisation current in air at low pressure, using a small part only of the range of the α -particles, is suggested.

The scattering of the α -particles in passage through air and

¹⁹ H. L. Bronson, *Phil. Mag.*, 1908, [vi], **16**, 291; *A.*, 1908, ii, 792.

²⁰ *Ann. Report*, 1906, 342.

²¹ Mlle. L. Blanquies, *Compt. rend.*, 1909, **148**, 1753; *Le Radium*, 1909, **6**, 230; *A.*, ii, 634.

²² H. Geiger, *Proc. Roy. Soc.*, 1909, **82**, A, 358.

metals²³ has been re-examined by the scintillation method.²⁴ In a good vacuum there is no scattering, but a single gold leaf is sufficient to cause notable scattering of the beam. It has also been established²⁵ that a diffuse reflection of α -rays occurs from the incident surface of a metal struck by α -rays, which is the greater the greater the atomic weight of the metal. Gold returns about twenty times as many α -particles as aluminium. In one experiment with platinum, about 1 in 8000 of the α -particles was reflected. The effect attains a maximum very quickly when the thickness of the reflector is increased, about half the maximum being attained by a thickness equivalent to only 2 mm. of air. Further search for any indication of the α -particle after it has passed its critical distance at which ionisation ceases²⁶ has resulted in failure. It has been shown that the positive charge transported by the particles and the secondary emission of electrons produced on impact cannot be detected beyond the critical distance.²⁷ It appears certain that the α -particle is quite suddenly stopped at the end of its detectable path, and becomes an ordinary atom of helium, but its kinetic energy at this point is still considerable, and may possibly furnish a source necessary for any process of atomic synthesis.²⁸

It is known that the heavier metals absorb the α -rays more strongly at the end of their path than initially.²⁹ The "air equivalent" of the metal, or thickness of air equivalent to a given thickness of metal, increases as the range increases. Paper and similar substances have air equivalents independent of the range, but in hydrogen the "hydrogen equivalent" increases for these substances as with the metals. The effects are cases of a general law that if the stopping power of any substance is taken as the standard, the relative stopping powers of substances of greater atomic weight than the standard increase as the range of the α -particle increases, while for substances of less atomic weight than the standard the converse is true.³⁰

The phenomenon of "initial recombination"³¹ shown exclusively by the ionisation produced by α -rays has received an elegant

²³ *Ann. Report*, 1906, 335.

²⁴ H. Geiger, *Proc. Roy. Soc.*, 1908, **81**, A, 174; *A.*, 1908, ii, 795.

²⁵ H. Geiger and E. Marsden, *Proc. Roy. Soc.*, 1909, **82**, A, 495; *A.*, ii, 782.

²⁶ *Ann. Report*, 1906, 335.

²⁷ E. Aschkinass, *Ann. Physik*, 1908, [iv], **27**, 377; *A.*, 1908, ii, 920; W. Duane, *Amer. J. Sci.*, 1908, [iv], **26**, 464; *Compt. rend.*, 1908, **146**, 958 and 1089; *A.*, 1908, ii, 553, 554.

²⁸ Compare W. H. Bragg, *Presidential Address: Australian Assoc.*, 1909.

²⁹ *Ann. Report*, 1907, 312.

³⁰ T. S. Taylor, *Amer. J. Sci.*, 1908, [iv], **26**, 169; *A.*, 1908, ii, 783; *Phil. Mag.*, 1909, [vi], **18**, 604; *A.*, ii, 850; *Physical Rev.*, 1909, **28**, 465.

³¹ *Ann. Report*, 1906, 338.

explanation. The α -particle leaves behind in its flight a long column of ions of cross section corresponding with that of the α -particle. Thus, in air at atmospheric pressure, the passage of an α -particle will leave about 10^5 pairs of ions crowded into 3 cm. length of one of these narrow columns. The real concentration of the ions in these columns is far greater than the average concentration for the whole gas, and recombination is favoured. Even a very intense electric field acting normally to the trajectories of the rays cannot completely prevent recombination, whereas it has been found that if the field acts at right angles to the trajectories the saturation current is far more readily and completely obtained.³² This seems to render unnecessary the earlier view that the ions generated from one atom were caught and prevented from escaping by other atoms in the same molecule.

β -Rays.

As is well known, the ratio e/m of the charge to the mass of the β -particle diminishes rapidly as the velocity (v) of the particle approaches that of light, and this has been generally explained by the increase in the (purely electromagnetic) mass of the electron at high velocity. The variation of e/m with v for the β -particles of radium has been redetermined³³ with great accuracy, and the various mathematical equations which have been proposed on theoretical grounds to express the law of variation have been thus subjected to experimental test. A grain of pure radium fluoride was placed in the centre of two parallel circular plates, 0.25 mm. apart, electrically charged to opposite sign. A photographic film on the inside of a cylinder coaxial with the plates and of twice their diameter received the trace of the deflected rays. A magnetic field acted in the direction parallel to one diameter of the plates. Thus it was ensured that only those rays escaped and reached the film for which the electrostatic deviation inside the plates exactly balanced the electromagnetic deviation. After the rays had escaped from the parallel plates, the magnetic force alone operated. If α is the angle between the direction of the rays and that of the magnetic field, and H and F are the strengths of the magnetic and electric fields respectively, $V = \frac{F}{H \sin \alpha}$, or, in words, each point

on the photographic trace corresponds with rays of a definite known velocity, fixed by the conditions of the experiment. The deviation of the trace at each point from the plane of the parallel plates

³² M. Moulin, *Compt. rend.*, 1909, **148**, 1757; *Le Radium*, 1908, **5**, 136; *A.*, 1908, ii, 921. The theory is ascribed to M. Langevin.

³³ A. H. Bucherer, *Physikal. Zeitsch.*, 1908, **9**, 755.

thus gives the magnetic deflection of homogeneous rays of known velocity, and so the variation of e/m with velocity is determined. As a result, it was established that $e/m_0 = 1.705 \times 10^7$, where m_0 represents the mass at low speeds, and $e/m = e/m_0(1 - \beta^2)^{1/2}$, where β is the ratio of the velocity of the particle to that of light. This is the Lorentz theoretical equation, and its experimental proof appears also to have important metaphysical consequences in establishing the Lorentz-Einstein principle of relativity.

Measurements³⁴ of the amount of negative electricity transported in a high vacuum from the bare active deposit of radium have led to the conclusion that as many electrons are emitted in the change of radium-*B* as in that of radium-*C*, and that for each α -particle from the latter there must be at least 20 and probably 50 electrons emitted. The electric and magnetic deflection of these electrons shows, however, that most of them move with the relatively feeble velocity of less than 4×10^8 cm. per second. They would thus possess very little penetrating power, and must be due to rays of the δ type, rather than to true penetrating β -rays. Over the distance of 0.5 mm. in air from the bare surface, however, they produce intense ionisation, comparable with that produced by the α -particles of radium-*C*. This perhaps explains the probably erroneous statement that radium-*B* emits α -particles.³⁵

With reference to the true β -particles emitted by radium-*C*, other experiments³⁶ have indicated that probably only one, or possibly two β -particles are emitted for each α -particle. The negative charge communicated in a high vacuum to an insulated brass cylinder, connected to an electrometer, from a glass tube, of wall thickness 0.078 mm., containing radium emanation, placed inside the cylinder, corresponded with 3.7×10^{10} β -particles per second (referred to 1 gram of radium), assuming the charge on each particle to be 4.67×10^{-10} E.S.U. Corrections for a small proportion of the β -particles of radium-*C* absorbed in the glass, and also for a small proportion of those from radium-*B* able to get through, gave the number 5.0×10^{10} (for radium-*C* alone). This correction involves, however, the unproved assumption that the total number of β -particles expelled from radium-*B* is equal to that from radium-*C*, and is therefore preliminary.

³⁴ W. Duane, *Le Radium*, 1908, 5, 71; *Amer. J. Sci.*, 1908, [iv], 26, 1; *A.*, 1908, ii, 748; E. Aschkinass, *Ann. Report*, 1907, 315; *Ann. Physik*, 1908, [iv], 27, 377; *A.*, 1908, ii, 920.

³⁵ *Ann. Report*, 1906, 346 and 347; F. A. Harvey, *Physikal. Zeitsch.*, 1909, 10, 46; *A.*, ii, 203 (also *Physical Rev.*, 1909); H. L. Bronson, *Physikal. Zeitsch.*, 1909, 10, 393; *A.*, ii, 634.

³⁶ W. Makower, *Phil. Mag.*, 1909, [vi], 17, 171; *A.*, ii, 204; *Brit. Ass. Rep.*, 1908, 601.

A fundamentally important consequence of the same experiments is the proof that the *number* of β -particles penetrating successive thicknesses of matter is reduced according to the same exponential law as when the absorption is determined in the usual way by measuring the ionisation the emergent particles can produce. The whole question of the nature of the β -rays, whether homogeneous or heterogeneous as regards velocity, and the exact meaning to be attached to their "absorption" in passage through matter,³⁷ is, in spite of numerous researches, still in a highly controversial state, and would scarcely repay very detailed discussion at the present stage.

In the first place, it has been established³⁸ that in passage through matter far more of the β -rays are scattered out of their original direction than are completely stopped, and relatively very thin screens—0.01 cm. of aluminium, or 0.002 cm. of gold—completely scatter a pencil of β -rays in all directions. Scattering alone can be represented by an exponential equation with a numerical "coefficient of scattering," which is for all substances about thirteen times greater than the true coefficient of absorption. This easy scattering is one of the many difficulties which make any exact theory of the nature of the absorption of β -rays difficult to establish.

In the second place, it appears that the so-called absorption-coefficient is very much influenced by relatively small differences in velocity. Thus, comparing the β -radiations of uranium-*X* and radium-*E*₂, the absorption-coefficients in aluminium are respectively 14 and 40 (cm.)⁻¹, whereas the velocities are, according to new determinations,³⁹ 2.76×10^{10} and 2.31×10^{10} cm. per second. Recent work to be considered has shown that if single β -ray products are dealt with, the absorption tends to be in almost all cases purely exponential, and since it is known that the number of β -particles is exponentially diminished, it follows that the velocity of the emergent particles is not altered by their passage through matter, for a diminution in velocity far too small to be directly detected by magnetic deflection methods would have a large influence in increasing the absorption-coefficient. Revolutionary conclusions, it is true, as to the heterogeneity of β -rays and their linear rather than exponential absorption by matter have been recently put forward, but it remains very open to question whether the methods employed were refined enough to bear such an interpretation.⁴⁰

³⁷ For a full discussion of the alternative possibilities at issue, see *Ann. Report*, 1907, 315.

³⁸ J. A. Crowther, *Proc. Roy. Soc.*, 1908, **80**, A, 186; *A.*, 1908, ii, 247; *Proc. Camb. Phil. Soc.*, 1909, **15**, 273; J. P. V. Madsen, *Phil. Mag.*, 1909 [vi], **18**, 909; *A.*, 1910, ii, 7.

³⁹ H. W. Schmidt, *Physikal. Zeitsch.*, 1908, **9**, 6.

⁴⁰ W. Wilson, *Proc. Roy. Soc.*, 1909, **82**, A, 612; O. Hahn and L. Meitner, *Physikal. Zeitsch.*, 1909, **10**, 948; *A.*, 1910, ii, 8; W. Wilson, *ibid.*, 1910, **11**, 101.

A working theory has been put forward, which, although almost certainly not rigidly true, has proved extraordinarily fruitful of new discoveries, according to which only one type of β -radiation is given out in each disintegration, and this radiation is strictly homogeneous as regards velocity and is absorbed by matter according to a purely exponential law. Departures of the β -ray absorption curves from this law indicate complexity of the β -rays, due to their being derived from more than one disintegration product.⁴¹ In order to avoid, as far as possible, complications due to "secondary" β -rays, aluminium is used as the absorbing material in taking the absorption curves, and the diameter of the electroscope is large compared with that of the pencil of rays to include also scattered radiation. In these circumstances, according to this theory, if the absorption curve of the β -rays is not exponential, a complex disintegration of more than one β -ray product is indicated, and in many cases where new β -ray products have been indicated in this way, they have subsequently been found to exist, and have been separated by new methods. It may be remarked in passing, however, that such a true exponential absorption applies at best only over a limited range. Thus, the penetrating β -radiation of uranium-*X*, from the first regarded as an example of homogeneous β -radiation, and for which even now no evidence of complexity has been advanced, is *not* exponentially absorbed by aluminium in any circumstances, the absorption-coefficient increasing continually and regularly to nearly three times the initial value before the rays are all absorbed.⁴² The advocates of the theory seem disposed to regard all such phenomena as due to unsuitable conditions of measurement, but it is to be noted that they have never yet shown a true exponential absorption curve except over a short range. It seems as though the β -particle probably does suffer some reduction in velocity in passage through matter, accounting for this change of the absorption-coefficient, although this diminution may well be too small to be detected directly by magnetic measurements.⁴³

It has also been found impossible completely to deviate the β -rays of either uranium or radium by magnetic fields several times as strong as is sufficient to deviate the great majority of the rays, and a very small proportion of the β -rays in each case seem to possess velocities very nearly indeed approaching that of light itself.⁴⁴ Nevertheless, it has been established for a great number of single β -ray products that pure exponential absorption of the

⁴¹ O. Hahn and L. Meitner, *Physikal. Zeitsch.*, 1908, 9, 321; *A.*, 1908, ii, 452.

⁴² M. Levin, *ibid.*, 1907, 8, 535; *A.*, 1907, ii, 836.

⁴³ H. W. Schmidt, *ibid.*, 1909, 10, 929; *A.*, 1910, ii, 7.

⁴⁴ F. Soddy, *Phil. Mag.*, 1909, [vi], 18, 858; *A.*, 1910, ii, 10.

rays can be obtained over short ranges by suitable methods of measurement, and only a few exceptions to the rule now remain to be cleared up.

In the following table, compiled from a mass of observations,⁴⁵ are collected the substances giving β -rays homogeneous in this sense, together with the thickness in mm. of aluminium necessary to cut down the β -rays to half-value.

Thorium- <i>A</i>	0.05	Radium- <i>B</i>	0.09
Thorium- <i>D</i>	0.441	Radium- <i>E</i> ₂	0.162
Uranium- <i>X</i>	{ 0.434	Radioactinium	0.04
	{ 0.012	Actinium- <i>A</i> less than	0.04
Radium.....	0.022	Actinium- <i>C</i>	0.24

A new short-lived product intermediate between meso-thorium and radio-thorium, known as thorium-2, also gives β -rays, which, however, appear complex. Radium-*C* gives probably two types of β -rays of not very different penetrating power, and a partial resolution of radium-*C* into two successive products has been effected. The list comprises many substances either new or not before suspected of giving β -rays, but in all such cases the β -radiation is of a very feeble penetrating power, which accounts for their either being wrongly ascribed to α -rays, or to their not being at first recognised as definite specific β -radiations. Whatever may be thought of the truth of the underlying conceptions, their fruitfulness in the discovery of new disintegration products cannot be denied. The further logical extension of this point of view, that each disintegration gives out one, and one only, definite homogeneous type of radiation—that when a substance gives out both α - and β -rays together, as, for example, radium itself and radium-*C*, they must be derived from successive changes—does not appear yet to be experimentally justified.

γ -Rays.

A great impetus has been given to the study of the γ -rays from the physical standpoint by the theory⁴⁶ that they also, like the β -rays, consist in the emission of discrete particles, rather than of pulses or waves of electromagnetic character similar to light. The theory, which is known as the neutral-pair theory, in that the discrete γ -particle is supposed to consist of an electrically neutral couplet composed of one negative and one, still itself unknown, positive electron, has hardly been seriously opposed so far as the γ -rays themselves are concerned. It is its further extension to the

⁴⁵ O. Hahn and L. Meitner, *Physikal. Zeitsch.*, 1908, **9**, 321, 649 and 697; *A.*, 1908, ii, 452, 920, 1007; 1909, **10**, 697 and 741; *A.*, ii, 849, 954; O. Hahn, *ibid.*, 1908, **9**, 246; *A.*, 1908, ii, 484.

⁴⁶ *Ann. Report*, 1907, 321.

X-rays, and ultimately possibly even to light itself, that has aroused opposition, and it is not difficult to foresee that the growth of the theory threatens to reopen fundamental questions of the nature of electromagnetic radiations in general and to modify the old undulatory theory, so as to bring it more into harmony with the modern views as to the discrete nature of the electric charge. A full consideration of the general question would be manifestly impossible here. The γ -rays only can be considered, although, as already remarked, this is to confine the evidence mainly to the side in favour of the newer theory. It is recognised that the γ -rays, directly, produce little, if any, detectable action. Their ionisation and other effects have been traced, at least mainly, to the secondary β -rays, into which they are in part transformed in passage through matter. On the new view a certain proportion of the neutral pairs is broken up on impact with atoms, the negative partner being liberated as a detectable β -particle. The main evidence in favour of the theory is that the γ -ray possesses momentum in the direction of its propagation. The secondary β -particles they generate move at first mainly in the direction of the original beam, whereas on the older pulse theory no such asymmetry is to be anticipated. The consequence is that there exists great disproportionality in the amount of secondary β -radiation from the incident and emergent faces of a plate traversed normally by a pencil of γ -radiation. It is easy to tell from this, without otherwise knowing, in which direction the γ -ray is moving, whilst on the older pulse theory this would be impossible.⁴⁷ A tendency from this point of view is to regard the β - and γ -radiation as mutually convertible in their passage through matter. The production of secondary γ -rays from β -rays has, however, not hitherto been observed,⁴⁸ although the existence of a feeble secondary γ -radiation produced by γ -rays has now been well established.⁴⁹ The interdependence of the primary β - and γ -radiation is questioned also on account of the great relative poverty in γ -rays, as compared to β -rays, shown by uranium as contrasted with radium.⁵⁰ Experiments on intensely active preparations of uranium-X, prepared from large quantities of uranium, have shown that the γ -rays possess, contrary to prior statements, only slightly less penetrating power than those from radium,⁵¹ but

⁴⁷ W. H. Bragg and J. P. V. Madsen, *Phil. Mag.*, 1908, [vi], **15**, 663; *A.*, 1908, ii, 556; **16**, 918; *A.*, ii, 112.

⁴⁸ H. Starke, *Ber. Deut. physikal. Ges.*, 1908, **10**, 267; *A.*, 1908, ii, 341.

⁴⁹ R. D. Kleeman, *Phil. Mag.*, 1908, [vi], **15**, 639; *A.*, 1908, ii, 553; A. S. Eve, *ibid.*, **16**, 224; *A.*, 1908, ii, 795; *ibid.*, 1909, **18**, 275; *A.*, ii, 783; J. P. V. Madsen, *ibid.*, **17**, 423; *A.*, ii, 365.

⁵⁰ F. Soddy and A. S. Russell, *Phil. Mag.*, 1909, [vi], **18**, 620; *A.*, ii, 851.

⁵¹ Compare *Ann. Report*, 1906, 352.

in proportion to the β -radiation they are over sixty times less powerful. The γ -rays are shown, with suitable methods of measurements, to be exponentially absorbed by matter (after a certain initial thickness), the absorption-coefficient having been redetermined for both radium and uranium γ -rays for a number of substances and shown to be nearly proportional to the density. Certain fruitful sources of confusion in previous measurements with regard to the numerical value of these coefficients have been cleared up.

A second and perhaps more essential modification brought about by the new theory concerns the source of energy of the secondary β - or cathode-radiation produced by γ -rays and X -rays respectively, in that it is ascribed to the energy of the primary beam, rather than to the internal energy of the atoms subjected to the radiation. The effects dealt with are in general complicated, more than one type of secondary radiation being produced. In the first place, the velocity of a part of the secondary β -rays produced by γ -rays appears to approximate to the velocity of the primary β -rays.⁵² This is in favour of the view that the β - and γ -rays are mutually convertible. In the second place, there is a tendency to explain the variations of secondary radiation with the atomic weight of the radiator,⁵³ without recourse to the view that any specific secondary radiation is emitted by the atoms of the radiator.⁵⁴

An earlier view, that the primary radiation acted as the trigger to release a certain internal store of energy from the atom in the form of secondary β -radiation, travelling with a velocity dependent on the nature of the atom rather than on that of the primary beam, is on its trial. It appears rather as if the character of the primary beam, and not at all that of the atom, decided the nature of the secondary radiation.

Thermo-Radioactivity.

The heat evolution of more than 1 gram of radium chloride, containing 0.7951 gram of radium (element) as determined by atomic weight estimations, has been redetermined. The conditions were somewhat indefinite as regards the proportion of the energy of the *penetrating* radiations converted into heat, the rays having to penetrate 1 mm. of glass and 5 mm. of copper before escaping from the calorimeter, so that all the β -radiation and a small part of the γ -radiation would be absorbed. The result was that 1 gram of

⁵² R. D. Kleeman, *Proc. Roy. Soc.*, 1909, **82**, A, 128; *A.*, ii, 364.

⁵³ *Ann. Report*, 1906, 350; 1907, 318.

⁵⁴ W. H. Bragg and J. P. V. Madsen, *Phil. Mag.*, 1908, [vi], **16**, 692; *A.*, 1908, ii, 921; J. P. V. Madsen, *ibid.*, 1909, **18**, 909; *A.*, 1910, ii, 7.

radium (element), in equilibrium with its products as far as radium-*C*, generates 118.0 gram-calories per hour. The uncertainty attaching to the amount of the products radium-*D* to radium-*F* in the preparation was not greater than 1/2 per cent.⁵⁵

A real advance in the measurement of minute evolutions of heat from radioactive preparations appears to have been made in the construction of a calorimeter capable of measuring the evolution of 0.001 gram-calorie per hour.⁵⁶ The apparatus consisted of two similar exhausted vessels, half full of ether, connected from below the ether-level by a capillary gauge containing a gas bubble, the motion of which was observed. The evolution of heat is recorded by the rise in the vapour pressure of the ether surrounding the preparation. The whole was elaborately insulated from external thermal influences, and the preparation to be tested was placed inside one of the vessels, together with a Peltier couple of iron-nickel, which was cooled by the passage of a current to neutralise exactly the heat evolved from the radioactive preparation. A preliminary determination with 0.8 milligram of radium chloride gave 120 calories per gram of radium per hour. A radio-thorium preparation gave 0.025 cal. per hour, which was of the same order as that from radium of similar activity. A polonium preparation gave 0.0117 cal. per hour. In the latter case, it was calculated that a quantity of radium giving the same ionisation current as the polonium would have given 0.011, so the result is favourable to the view that the heat disengaged is almost entirely due to the kinetic energy of the α -particles expelled.

A determination of the heat generated by thorium oxide has given as the result that 1 gram, in equilibrium with its products of disintegration, gives 2.1×10^{-5} calories per hour, which is 3.2×10^6 times less than that given by radium in equilibrium.⁵⁷ Experiments have also been made on pitchblende, but final results are not yet to hand.⁵⁸ The earlier observation that the absorption of X-rays in lead produces more heat than in zinc has now been definitely shown to be erroneous.⁵⁹

⁵⁵ E. v. Schweidler and V. F. Hess, *Sitzungsber. K. Akad. Wiss. Wien*, 1908, **117**, (iia), 879.

⁵⁶ W. Duane, *Compt. rend.*, 1909, **148**, 1448 and 1665; *A.*, ii, 534, 637.

⁵⁷ G. B. Pegram and H. W. Webb, *Physical Rev.*, 1908, **27**, 18.

⁵⁸ Compare J. Joly, *Radioactivity and Geology*, p. 11; H. H. Poole, *Phil. Mag.*, 1910, [vi], **19**, 314. The result is considerably higher than is to be anticipated from the simple disintegration theory.

⁵⁹ *Ann. Report*, 1906, 351; H. A. Bumstead, *Phil. Mag.*, 1908, [vi], **15**, 432; *A.*, 1908, ii, 342.

Recoil of Disintegration Products.

When an atom of mass of the order of 200 expels an α -particle of mass 4 with a velocity of the order of 2×10^9 cm. per second, it follows, if the expulsion obeys ordinary mechanical laws, that the product of the disintegration must recoil with a velocity of the order of 4×10^7 cm. per second in the direction opposite to that of the expelled α -particle. Conclusive evidence of this recoil has now been obtained, and the phenomenon, in addition, has been the means of adding an extremely valuable new method for the separation of radioactive products, too difficult to be effected by ordinary chemical and physical processes. In the last Report, attention was directed to the feeble residual activity left after the active deposit of actinium produced by the actinium emanation completely decays, which was described as one of the most interesting radioactive phenomena of the time.⁶⁰

With very active preparations of radio-actinium this activity has been further studied, and it has been shown to be due to actinium-*X*, which is the parent rather than the product of the active deposit. Now actinium-*X* is not volatile even at a red-heat. The residual activity is completely prevented when the actinium preparation is wrapped in tissue paper, which allows the emanation to go through, and the ordinary active deposit of actinium to be formed without hindrance. Actinium-*X*, free from its parent radio-actinium, does not show the residual activity, although, of course, it generates emanation, and produces the ordinary active deposit. The explanation is that it is only produced by nascent actinium-*X*, which recoils from the actinium preparation at the moment of its formation from radio-actinium by the expulsion of the α - and β -particles.⁶¹

It is found that a negative charge on the plate being made active increases greatly the amount of recoiled actinium-*X* collected. In the case of a long known similar phenomenon with radium-*B*,⁶² which is not "volatile" in the ordinary sense, but appears so at the moment of its formation from radium-*A* by expulsion of an α -particle, the recoil explanation had already been suggested. This case and many others⁶³ has been reinvestigated, and been found parallel to that of actinium-*X*, the amount of radium-*B* collected being greatly assisted if the collecting surface is negatively charged with respect to the active deposit. It appears that the recoiled

⁶⁰ *Ann. Report*, 1907, 328.

⁶¹ O. Hahn, *Physikal. Zeitsch.*, 1909, 10, 81; *A.*, ii, 206.

⁶² Miss Brooks, see Rutherford's *Radioactivity*, 2nd edition, p. 392.

⁶³ O. Hahn and L. Meitner, *Ber. Deut. physikal. Ges.*, 1909, 11, 55.

product of disintegration carries a positive charge, as it has been found to do in other cases. In the actinium active deposit itself, it is known that the first substance, actinium-*A* (half-period, thirty-six minutes), only expels feeble β -radiation and no α -radiation, whereas the product, actinium-*B*, expels an α -particle (possibly two in rapid succession)⁶⁴ in its rapid change (half-period, 2.15 minutes) into actinium-*C*. This latter substance, which had been discovered shortly before these experiments, as it is separated from a solution of the active deposit by adsorption with charcoal and finely divided metals,⁶⁵ gives only the penetrating β - and γ -rays of actinium, and has a half-period of 5.1 minutes. It was found that pure actinium-*C*, decaying exponentially to zero with a period of 5.1 minutes, and giving only β - and γ -radiation, could be obtained by exposing a negatively charged surface in the neighbourhood of the active deposit. In suitable circumstances, more than half the theoretical production of pure actinium-*C* could be got in this way. In these rapid successive changes the new method of separation is a most powerful new weapon of attack. Changes too rapid for any other process to be of service can thus be detected. The next result was new. Thorium-*A* (half-period, 10.6 hours) gives no α -particle whereas the product thorium (*B* and *C*) (half-period, 60.4 minutes⁶⁶ gives two of different range, and it was previously thought the β - and γ -rays also. The recoil method, however, yielded from the thorium active deposit an entirely new product, called thorium-*D*, giving only the β - and γ -rays, decaying exponentially to zero with half-period 3.1 minutes. It gave no α -rays on decaying, and hence must follow thorium (*B* and *C*) in the series. The absorption curves of the thorium-*D* β -rays are identical with those of the more penetrating of the two types given by the thorium active deposit. An independent set of similar experiments, carried out with a condensed film of radium emanation in a vacuum,⁶⁷ showed that radium-*A*, -*B*, and -*C* recoiled from the emanation. No electric field was employed in these experiments, and it was found that the amount recoiled varied inversely as the square of the distance, and was very markedly diminished by the presence of air or other gases. The air absorbed the products recoiled exponentially, and in one case, under 0.44 mm. pressure with 6.5 cm. of path, half was absorbed by the air. Hydrogen was found to absorb the recoil products far more than air in proportion to its density.

In both sets of investigation it was established that the expulsion of the β -particle by itself produces a detectable recoil of the

⁶⁴ Compare Mlle. Blanquies, p. 236.

⁶⁵ O. Hahn and L. Meitner, *Physikal. Zeitsch.*, 1908, **9**, 649; *A.*, 1908, ii, 920.

⁶⁶ *Ann. Report*, 1906, 342.

⁶⁷ S. Russ and W. Makower, *Proc. Roy. Soc.*, 1909, **82**, A, 205; *A.*, ii, 455.

product, which is far smaller than that produced by the α -particle. Thus, the active deposit of radium, after being left long enough for the rapidly changing α -ray substance, radium-*A*, completely to disintegrate, exposed in a vacuum gave a recoil product of radium-*C* alone, which could only be due to the β -particle expelled in the change of radium-*B*. Further investigations have established that radium-*C* itself gives a new rapidly decaying recoil product.⁶⁸ The complex nature of radium-*C* has already been adumbrated on several grounds.⁶⁹ In addition to the evidence drawn from the very penetrating character of the α -radiation, which indicates a far more rapidly changing product than that ascribed to radium-*C* (half-period, 19 minutes), and to the discrepancies between the calculated periods of radium-*D*, the substance gives a complex β -radiation not exponentially absorbed by aluminium (compare p. 240). Recoil experiments with pure radium-*C* itself, separated by chemical methods from a solution of the active deposit of radium, gave products decaying very rapidly, with a half-period of from 1 to 2.5 minutes. Only very feeble recoil effects were obtained, indicating that radium-*C*₁ gives β -rays and no α -rays. The recoil product, radium-*C*₂, gave α -rays, and decayed exponentially with a period of about two minutes.

There is thus evidence that radium-*C* is complex and consists of two rapidly succeeding products, whilst three are suggested. The results so far, however, are not very conclusive. The new method may perhaps also apply to cases like radium-*F*, to determine whether it is the true end-product of the series. There is little doubt that the new method may serve to throw light on problems capable of being attacked in no other way.⁷⁰ There is some evidence that radium-*F* gives a recoil product which carries a positive charge, and is detectable thereby. If so, a method may exist for the recognition of the final end-product of the series, even although it is completely non-radioactive. Even the determination of the atomic weight by measurements of the magnetic and electric deflexion of the charged particle is not entirely out of the question.

Influence of Temperature on Radioactive Change.

This subject has been left in an unsatisfactory state in previous reports, and, in spite of further researches, still remains unsettled, a real conflict existing in the experimental results of different workers, and even of the same workers in different experiments.

⁶⁸ O. Hahn and L. Meitner, *Physikal. Zeitsch.*, 1909, **10**, 697; *A.*, ii, 849.

⁶⁹ *Ann. Report*, 1907, 312 and 338.

⁷⁰ J. C. McLennan, *Nature*, 1909, **80**, 490; compare also E. Aschkinass, *Ann. Physik*, 1908, [iv], **27**, 377; *A.*, 1908, ii, 920.

Many of the results already recorded⁷¹ have been independently confirmed.⁷² The β - and γ -activity of radium-C from a quartz tube containing the radium active deposit has now been measured during the actual heating. Soon after heating to 1100° or 1200°, a decay more rapid than the normal occurred, gradually becoming normal as the heating was maintained. On cooling, the decay was at first less than, but increased to, the normal. Disturbances due to unequal volatilisation of the products and to the permeability of the tube at high temperature appear to have been eliminated. With the emanation itself in the quartz tube, a rapid increase at first was always observed. In the first experiments, cooling brought the activity below the normal, and the activity then gradually increased to the normal. In later experiments, however, this increase was not observed. This was attributed to non-homogeneity of the emanation, for which some other as yet incomplete evidence exists, the consideration of which may profitably be postponed.⁷³ With the hard γ -rays little change was observed. It is concluded that the changes both of the emanation and radium-A are accelerated by rise of temperature. Other investigators continue to deny any effect of temperature.⁷⁴ The changes, in any case, are small, but the evidence in their favour cannot be altogether ignored. The question remains an open one.

Helium and Ultimate Disintegration Products.

A measurement of the rate of production of helium from 0.07 gram of radium chloride, by absorbing condensable gases in charcoal cooled in liquid air (sometimes in liquid hydrogen), and measuring the increase in the amount of residual gas in a compression gauge, gave 0.37 cu. mm. of helium per gram of radium per day, which is very nearly the theoretical value (compare p. 235).⁷⁵ The radium was heated to 450° before the measurements, and the purity of the gas measured as helium was established. The production of infinitesimal quantities of helium from uranium and thorium has been established by direct experiments with large quantities of materials by means of the calcium absorption process, which ensures that the helium could not have been of atmospheric

⁷¹ *Ann. Report*, 1907, 336.

⁷² W. Engler, *Ann. Physik*, 1908, [iv], **26**, 518; *A.*, 1908, ii, 650.

⁷³ Compare also Rutherford and Tuomikoski, *Mem. Manchester Phil. Soc.*, 1909, **53**, xii.; *A.*, ii, 456; Rutherford, *Phil. Mag.*, [vi], 1909, **17**, 723; *A.*, ii, 456; A. Debiérne, *Compt. rend.*, 1909, **148**, 1264; *A.*, ii, 534.

⁷⁴ H. W. Schmidt, *Physikal. Zeitsch.*, 1908, **9**, 113; *A.*, 1908, ii, 141; Makower and Russ (controversial against the latter), *ibid.*, 250; *A.*, 1908, ii, 449; H. W. Schmidt and P. Cermak, *ibid.*, 816.

⁷⁵ Sir J. Dewar, *Proc. Roy. Soc.*, 1908, **81**, A, 280; *A.*, 1908, ii, 921.

origin. The amount produced in both cases is of the same order as the theoretical. In the uranium experiments a spectroscopic estimation of the rate of production agreed with the theoretical on the assumption that each atom of uranium disintegrating gives one, not two, atoms of helium, but the estimate is necessarily a minimum one.⁷⁶ The amount corresponds with only 11 cu. mm. per thousand kilograms of uranium per year.

Attempts have been made to use the amount of helium contained in common rocks and minerals as a criterion of their geological age. Except in the one definite case of beryl, which contains a marked amount of helium and practically no radioactive matter, and possibly of the saline minerals, such as carnallite, the amounts of helium correspond roughly with what is to be expected from the amount of uranium (measured as radium) and thorium.⁷⁷ It has been established, however, that under laboratory conditions the rate of escape of helium, even from dense, compact minerals like thorianite, far exceeds the possible rate of production, so that the primitive natural conditions must have been more favourable to the retention of helium.⁷⁸ This escape, of course, would invalidate the estimation of geological age, making it too low. However, the most recent results with hæmatites of various geological horizons, from Oligocene to Devonian, indicate ages far greater than are usually ascribed by geologists.⁷⁹ Still more recent results with zircons of geological ages from Tertiary to Archean, indicate a very close agreement between the age and the ratio of helium to radioactive matter. Doubt has been thrown on the existence of argon in zirconium minerals, such as malacone, except in spectroscopic quantities.⁸⁰ Doubt has also been thrown on the possibility of lead being the ultimate product of the uranium disintegration,⁸¹ owing to the absence (less than 0.01 per cent.) of lead in a fine crystal of autunite (hydrated uranium calcium phosphate). Independent spectroscopic examination of another specimen of this mineral, it is true, and on Cornish tobernite (hydrated uranium copper phosphate), showed lead as the only

⁷⁶ F. Soddy, *Phil. Mag.*, 1908, [vi], **16**, 513; *A.*, 1908, ii, 921; *Physikal. Zeitsch.*, 1909, **10**, 41; *A.*, ii, 207; *Proc. Phys. Soc.*, 1909, 178.

⁷⁷ R. J. Strutt, *Proc. Roy. Soc.*, 1908, **80**, A, 572; **81**, A, 272 and 278; *A.*, 1908, ii, 649, 922, 923; J. W. Waters, *Phil. Mag.*, 1909, [vi], **18**, 677; *A.*, ii, 848.

⁷⁸ R. J. Strutt, *Proc. Roy. Soc.*, 1909, **82**, A, 166; *A.*, ii, 457; J. A. Gray, *ibid.*, 301; *A.*, ii, 570.

⁷⁹ R. J. Strutt, *ibid.*, 1909, **83**, A, 97; *A.*, 1910, ii, 9; *ibid.*, meeting of 9/12/09, abstract only available.

⁸⁰ R. J. Strutt, *ibid.*, 1908, **80**, A, 572; *A.*, 1908, ii, 649; C. F. Hogley, *Phil. Mag.*, 1909, [vi], **18**, 672; *A.*, ii, 884; A. von Antropoff, *Zeitsch. Elektrochem.*, 1908, **14**, 585; *A.*, 1908, ii, 943.

⁸¹ W. Marckwald and B. Keetman, *Ber.*, 1908, **41**, 49; *A.*, 1908, ii, 144.

important foreign constituent. The quantity in the first case was, however, only such as would be formed theoretically in 10,000 years.⁸² The mineral must either be of extremely recent formation, or lead cannot be the ultimate product. With regard to the ultimate product of the thorium series, the active deposit of thorium appears to decay away completely, leaving no feeble residual activity, and the unknown product of the last known member, thorium-*D*, if not entirely stable, must possess a life period of at least two million years.⁸³ In this research the periods of thorium-*A* and -*B* were redetermined very accurately, and found to be 10·605 hours and 60·4 minutes respectively.

The Radioactive Emanations.

Physical Properties of Radium Emanation.—Several workers now having a considerable fraction of a gram of pure radium, many investigations have been carried out on the properties of pure radium emanation. The volume of the emanation in equilibrium with 1 gram of radium has been found to be almost exactly that theoretically deduced (p. 235) on the assumption that one mono-atomic molecule of emanation results per atom of radium disintegrating, the most trustworthy measurements agreeing in giving about 0·58 to 0·6 cu. mm. at N.T.P. The initial rapid contraction of volume of freshly separated emanation, although not entirely explained, has been traced to impurities, which have in no case been entirely eliminated. The measurements have therefore been taken of the minimum volume reached after a few hours. The amount of emanation measured has been compared by γ -ray measurements with a standard radium preparation.⁸⁴ The volume of emanation has been proved to be proportional to its γ -activity, independently of the time of its accumulation.

On this infinitesimal quantity of gas, determinations have been made of the spectrum, vapour pressure, boiling point, freezing point, critical temperature, critical pressure, and the density of the liquid. The spectrum⁸⁵ is now fairly well known, having been photographed both with the prism and grating with sufficiently concordant results. An unexplained fact is the presence of xenon

⁸² J. A. Gray, *Phil. Mag.*, 1909, [vi], **18**, 816 and 937; *A.*, ii, 956.

⁸³ F. v. Lerch, *Sitzungsber. K. Akad. Wiss. Wien.*, 1907, **116**, (iia), 1443.

⁸⁴ E. Rutherford, *Phil. Mag.*, 1908, [vi], **16**, 300; *A.*, 1908, ii, 791; R. W. Gray and Sir W. Ramsay, *Trans.*, 1909, **95**, 1073; A. Debierne, *Compt. rend.*, 1909, **148**, 1264; *A.*, ii, 534.

⁸⁵ E. Rutherford and T. Royds, *Phil. Mag.*, 1908, [vi], **16**, 313; *A.*, 1908, ii, 787; T. Royds, *ibid.*, 1909, [vi], **17**, 202; *A.*, ii, 206; A. T. Cameron and Sir W. Ramsay, *Proc. Roy. Soc.*, 1908, **81**, A, 210; *A.*, 1908, ii, 786; T. Royds, *ibid.*, 1909, **82**, A, 22; *A.*, ii, 287; H. E. Watson, *ibid.*, 1909, **83**, A, 50; *A.*, ii, 954.

in the emanation spectrum on one occasion, for which no source has been suggested. The vapour-pressure curve of the emanation has been plotted in one set of investigations from 500 mm., 202.6° Abs. (solid) to 47,450 mm., 377.50° Abs. (critical temperature), the freezing point being given at -71°. ⁸⁶ The liquid emanation observed in a microscope is described as at first colourless and transparent like water, but as its temperature is reduced it ceases to transmit light, and on further cooling with liquid air the solid emanation glows with great brilliancy, at first like a small steel-blue arc lamp, whilst at a still lower temperature the glow is a brilliant orange-red. From a comparison of its boiling point, critical temperature, and critical pressure with those of the other inert gases, it was concluded that the atomic weight of the emanation was 176. The theoretical value, on the assumption that only an α -particle is expelled in the production of the emanation from radium, is 222.5, which has also been advocated from the same data on other grounds. ⁸⁷ Both numbers would fit in the Periodic Table equally well. Before this evidence from the physical constants can be considered beyond criticism, it would be of interest to have similar measurements carried out with 0.1 cu. mm. of xenon, krypton, and argon.

Diffusion-Coefficients.—The coefficients of diffusion of all three emanations have been redetermined with considerable accuracy, but the deduction therefrom of the molecular weight of the emanation appears to be beset with uncertainty. In the first place, the effect of a charge on the molecule, which at least initially it may reasonably be supposed to have, has been considered to be a disturbing factor sufficient completely to vitiate the method and to give too low values for the molecular weight. ⁸⁸ Possibly another disturbing factor, which also would apply mainly to the short-lived emanations, is the effect of recoil, causing the emanation molecule at first to move more rapidly than that of an ordinary gas of similar molecular weight and temperature. For the long-lived radium emanation, however, one would not expect either objection to apply, unless, indeed, the emanation molecule has its charge maintained by the ions in the gas. It has been suggested that fresh evidence might be obtained from a comparison of the thermal conductivities of the emanation and the heavier argon gases at very low pressure, ⁸⁹

⁸⁶ R. W. Gray and Sir W. Ramsay, *loc. cit.*; compare also E. Rutherford, *Phil. Mag.*, 1909, [vi], 17, 723; *A.*, ii, 456.

⁸⁷ G. Rudolf, *Zeitsch. Elektrochem.*, 1909, 15, 749; *A.*, ii, 954.

⁸⁸ Sir J. J. Thomson, Camb. Phil. Society's Meeting, 8/11/09. Abstract only so far available.

⁸⁹ F. Soddy and A. J. Berry, Royal Soc. Meeting, 9/12/09; F. Soddy, *Nature*, 16/12/09.

which have been shown with argon and neon to agree fairly well with the values calculated from the kinetic theory.

The most notable experimental advance with the radium emanation has been the direct comparison of its velocity of diffusion in hydrogen through asbestos plugs with that of mercury vapour at 250° and 275°. This is the first time direct comparison has been made with a monatomic gas of similar molecular weight.⁹⁰ The coefficients of diffusion of the emanation at the two temperatures were 0.0341 and 0.0376, as compared with 0.0370 and 0.0407 for mercury. On the assumption that the molecular weights are proportional to the squares of the diffusion-coefficients, the molecular weight of the emanation is 234.5. Another determination⁹¹ in air at the ordinary temperature has been made by Curie and Danne's original method, with improvements, by measuring the exponential escape of emanation from a bulb provided with a long, narrow tube. This gave the coefficient of diffusion 0.1015 at N.T.P. If the same law holds for the emanation as for hydrogen, water, and carbon dioxide, the molecular weight would be between 70 and 100, but no monatomic gas seems to have had its coefficient of diffusion so measured. The coefficient of diffusion of the actinium emanation has been deduced from the exponential decrease, with height, of the active deposit on a vertical wire in a long cylinder, at the bottom of which an actinium preparation was spread.⁹² Also the rate of diffusion of actinium and thorium emanations have been compared by the same method.⁹³ In the first investigation the coefficient was determined in air at various pressures, and in carbon dioxide and hydrogen. It was found that the ratios of the diffusion-coefficients in air to those in hydrogen and carbon dioxide respectively were the same for the actinium emanation as for ether, alcohol, water, etc. It was concluded that the molecular weight must be about 70 whatever the conditions under which diffusion is measured. In the second investigation, the values found for the actinium emanation agreed well with those calculated by Graham's law from the diffusion-coefficients in air when diffusion was carried out in hydrogen, carbon dioxide, sulphur dioxide, and argon. The actinium emanation coefficient was 1.19 times that of the thorium emanation (in air at 76 cm.), and it was concluded that the molecular weight of the thorium emanation is 1.42 times that of actinium. This question of the molecular weights of the emanation is thus still far from settled, and is one of the greatest importance in the present state of the subject.

⁹⁰ P. B. Perkins, *Amer. J. Sci.*, 1908, [iv], 25, 461; *A.*, 1908, ii, 552.

⁹¹ L. Chaumont, *Le Radium*, 1909, 6, 106; *A.*, ii, 781.

⁹² C. Bruhat, *Compt. rend.*, 1909, 148, 629; *Le Radium*, 1909, 6, 67; *A.*, ii, 300.

⁹³ S. Russ, *Phil. Mag.*, 1909, [vi], 17, 412; *A.*, ii, 366.

Condensation.—An interesting, but somewhat incomplete, investigation⁹⁴ has shown that whereas the temperature of condensation of the radium emanation in tubes of copper, iron, tin, and silver is about the same (-155°) as that originally found, in glass the temperature is about 20° lower. In silvered glass the temperature was little, if any, higher than in ordinary glass. This may explain the differences between the initial experiments, in which it was found that the temperature of condensation (in metal tubes) was extremely sharp, and those of a large number of later investigators (working with glass apparatus), who have found that by continued pumping some emanation can be continuously extracted even when it is kept at liquid-air temperature. Investigations have also been carried out on the absorption of the emanations by charcoal and other porous materials.⁹⁵ Not much hitherto has been known of the condensation of the actinium emanation, as its rapid decay (to 1/2 per cent. in 30 seconds) makes the experimental work difficult. A very complete and careful comparison⁹⁶ has now been made between its behaviour at low temperature and that of the thorium emanation, with the notable result that the two emanations are so completely alike as regards their condensation that they can barely be distinguished. This is unexpected in view of large difference in molecular weight indicated by diffusion experiments. At -120° both emanations begin to condense, and condensation is complete at -150° . If anything, the actinium emanation is slightly the more volatile. The disengagement of the emanation from radium-barium chloride and fluoride has been further studied in great detail. In the case of the former, the temporary minimum of emanating power passed through at 920° , 25° below the melting point, has been associated with a point of polymorphic transformation.⁹⁷ Radium salts precipitated along with hydrates of the rare earths have a very strong emanating power in the cold in the dry state, as in the case of the actinium preparations.⁹⁸ The great loss of emanation from dry radium bromide in the cold, as compared, for example, with radium chloride, is perhaps to be ascribed to the greater spontaneous instability of the bromide, and its rapid conversion into hydroxide and carbonate by the action of the air.

The Question of Transmutational Reactions Produced by the

⁹⁴ A. Laborde, *Le Radium*, 1909, **6**, 289; *A.*, ii, 634.

⁹⁵ Compare also E. Henriot, *ibid.*, 1908, **5**, 41; *A.*, 1908, ii, 651; R. W. Boyle, *Phil. Mag.*, 1909, [vi], **17**, 374. (See *A.*, 1908, ii, 1005.)

⁹⁶ S. Kinoshita, *ibid.*, 1908, [vi], **16**, 121; *A.*, 1908, ii, 652.

⁹⁷ *Ann. Report*, 1907, 332; L. Kolowrat, *Le Radium*, 1909, **6**, 321; *A.*, 1910, ii, 91.

⁹⁸ H. Herschfinkel, *Compt. rend.*, 1909, **149**, 275; *A.*, ii, 714.

Radium Emanation.—The experiments on the presence of neon with helium in the gases produced by the action of radium emanation upon liquid water⁹⁹ have been repeated by the original investigators, who conclude that they furnish conclusive evidence of the first observed case of transmutation.¹ In the experiment described, the analysis given showed that nearly 0.3 c.c. of nitrogen gas was present. Other workers have carried out similar experiments, and obtained in four out of five experiments no indication of neon in the helium produced. It was found that when 1/10 c.c. of air was added to the helium, and the gas again subjected to cold charcoal, the neon spectrum could be clearly seen. In the fifth experiment neon was observed, which was ascribed to atmospheric origin through air leakage.² The presence of lithium in salts of copper, subjected to the action of the radium emanation, was not observed in a series of independent experiments carried out with very carefully purified materials in platinum vessels.³ It was shown that water originally free from lithium, after standing in glass vessels for a short time, contained it in detectable quantities. The presence of lithium in opaque and transparent fused quartz was also detected. The occurrence of lithium in radioactive minerals has been examined. Lithium in spectroscopic quantity was found in all, but no relation existed between the amount of lithium and that of copper. Autunite contains no copper, and carnotite but little copper, and relatively much lithium.⁴ Experiments to detect lithium after the direct action of the rays and emanation of radium on salts of copper and gold have given negative results.⁵

The formation of considerable quantities of carbon dioxide by the action of carbon dioxide-free radium emanation on solutions of hydrofluosilicic acid, titanium sulphate, zirconium nitrate, thorium nitrate, and lead chlorate, and its absence in similar experiments with mercurous nitrate, has been interpreted on the view that the elements of the carbon group are degraded into carbon by the action of the emanation, the more readily (except lead) the higher the atomic weight of the element.⁶ From the thorium solution, 2.9 milligrams of carbon, as carbon dioxide, were obtained per cu. mm. of emanation employed.

⁹⁹ *Ann. Report*, 1907, 335.

¹ Sir W. Ramsay and A. T. Cameron, *Trans.*, 1908, **93**, 992.

² E. Rutherford and T. Royds, *Phil. Mag.*, 1908, [vi], **16**, 812; *A.*, 1908, ii, 1006.

³ Mme. Curie and Mlle. Gleditsch, *Compt. rend.*, 1908, **147**, 345; *A.*, 1908, ii, 793.

⁴ Mlle. Gleditsch, *ibid.*, **146**, 331; *A.*, 1908, ii, 246; Sir W. Ramsay and A. T. Cameron, *ibid.*, 456; *A.*, 1908, ii, 247.

⁵ E. P. Perman, *Trans.*, 1908, **93**, 1775.

⁶ Sir W. Ramsay and F. L. Usher, *Ber.*, 1909, **42**, 2930; *A.*, ii, 850.

The Radioactive Decomposition of Water and Various Gases.

For six or seven years the source of the excess of hydrogen always present in the electrolytic gas evolved in the radioactive decomposition of water has remained unaccounted for, but much fresh light has recently been thrown on the question. Contrary to earlier observations, the decomposition of water by the penetrating rays only of a considerable quantity of radium has been observed.⁷ After the first three days, for some months, a steady evolution of gas occurred, followed by a slow progressive diminution in the rate of production. The steady evolution was 0.115 c.c. per day per gram of radium, which is about 1 per cent. of the amount evolved from radium in solution. On testing the gas, it was found to be pure hydrogen, and an amount of hydrogen peroxide was found in the water nearly corresponding with the equation:

It is calculated that rather more than 0.01 per cent. of the total energy evolved by the radium can be utilised in this reaction. No such effect was observed for X-rays, but ultraviolet light from a quartz mercury vapour lamp brings about the same reaction. This is in conformity with the observation that hydrogen peroxide is found in greater amount in snow and rain during the daytime than at night, and that it is not found in dew.

An unaccountable cessation in the normal evolution of hydrogen and oxygen from a solution containing more than 0.2 gram of radium has been recorded. After some months of normal gas evolution, very little gas was generated for two months, and then the normal rate of evolution (25 c.c. per week) was resumed and maintained.⁸ As a result of a long series of experiments on the chemical action of the radium emanation on water, mixtures of hydrogen and oxygen, both moist and dry, oxides of carbon, hydrogen chloride, ammonia, and mixtures of nitrogen and hydrogen, it was found that the general law could be formulated that each molecule of emanation disintegrating produces a definite effect. The volume changes in the gases brought about by the presence of a definite quantity of emanation proceed exponentially with the time, the constant, for the average of all the experiments, being the same as that of the decay of the radium emanation.⁹ Liquid water is decomposed, whereas the mixed gases, hydrogen

⁷ A. Debierne, *Compt. rend.*, 1909, **148**, 703; *Le Radium*, 1909, **6**, 65; *A.*, ii, 364; M. Kernbaum, *Compt. rend.*, 1909, **148**, 705; **149**, 116 and 273; *Le Radium*, 1909, **6**, 225; *A.*, ii, 364, 714, 717.

⁸ R. W. Gray and Sir W. Ramsay, *Trans.*, 1909, **95**, 1073.

⁹ Sir W. Ramsay and A. T. Cameron, *ibid.*, 1908, **93**, 966; compare also H. Herschfinkel, *Le Radium*, 1909, **6**, 228. (See *A.*, ii, 778.)

and oxygen, in presence of phosphoric oxide are recombined. Carbon dioxide is decomposed with deposition of carbon and formation of carbon monoxide, whilst the latter gas also deposits carbon and forms carbon dioxide and some oxygen. Hydrogen chloride and ammonia are decomposed into their constituents, whilst mixtures of nitrogen and hydrogen contract in presence of fused calcium chloride, which absorbs ammonia. At 130° mixtures of hydrogen and oxygen recombine, whereas the reverse change with steam did not appear to take place. The general action of radioactive substance in causing chemical combination appears to be well explained by dissociation of molecular aggregates, and the temporary formation of free atoms, rendering the substances extremely reactive, as in the so-called nascent state. This great chemical reactivity of otherwise inert substances in presence of the radioactive materials has always to be remembered in the interpretation of their actions.

The Active Deposits.

The conditions of the deposition of the active deposit, with and without the action of electric fields, and of gravity¹⁰ have been examined in great detail. The amount of active deposit attracted to the negative electrode in the radium emanation varies with the voltage in a manner closely analogous to the ionisation current through the gas, indicating that the specific velocity and coefficient of recombination is of the same order of magnitude for the active matter as for the ions in the gas. Hence the practically important result follows that to get the maximum deposition a voltage sufficient to produce saturation in the gas is necessary in each case.¹¹ Without the action of an electric field or gravity, by working with vertical parallel plates at varying distances apart, a maximum distance apart exists beyond which the amount of active deposit does not increase, indicating that the time taken for the particles to diffuse to the plates exceeds their period of average life. This limiting distance apart, however, is attained at a distance twelve times less than indicated by theoretical considerations, as though the particles of the active products were aggregations very much larger than single molecules.¹² It has been established that radium-A is absent from that part of the active deposit falling under gravity, the time taken in falling being greater than the short life-period of this substance. This was done very neatly by exposing two horizontal superimposed

¹⁰ *Ann. Report*, 1907, 336.

¹¹ H. W. Schmidt, *Physikal. Zeitsch.*, 1908, 9, 184.

¹² A. Debierne, *Le Radium*, 1909, 6, 97.

plates to the emanation, and then, after the removal of the plates from the emanation, balancing the ionising current from the under surface of the upper plate against that due to the upper surface of the lower plate. The net effect was due to the gravity-deposited activity alone, and its decay curve showed no rapid initial drop characteristic of radium-*A*.¹³ By finding the maximum distance between the plates beyond which the gravity-deposited activity did not further increase, the time of fall and, from Stokes' law, the size of the falling particle were estimated. In moisture-laden air, the size found was much greater than in dry, and reached diameters from 150 to 280 $\mu\mu$, which is well within the region of ultramicroscopic vision. Supporting this inference, the formation of persistent mists has been observed in flasks containing moist, but not necessarily saturated, air illuminated by the arc. These last sometimes over a month, and gradually disappear as the emanation decays. They have been traced to chemical actions, such as the formation of oxides of nitrogen from the air, and the oxidation of sulphur from india-rubber stoppers, and are especially dense in chemically reactive gases such as chlorine.¹⁴ The results seem to have a bearing on the importance of radioactivity in producing meteorological phenomena. By experiments on the activity deposited on rods immersed in the emanation at various temperatures, it has been found that at 900° neither radium-*A* nor -*B* will deposit, whereas radium-*C* will, and, since the deposition under these conditions is uninfluenced by the electric field, it is concluded that radium-*C* is not charged at the moment of its formation from radium-*B*. The following volatilisation points are given: radium-*A*, 900°; radium-*B*, 600°; radium-*C*, 1200°.¹⁵ A full comparison of the behaviour of the active deposits from the three emanations in an electric field in gases at various pressures has been carried out.¹⁶ The conclusion is come to that some of the particles of the active deposit carry *negative* charges, for, especially in the case of actinium, some of the deposit is attracted to the positive electrode. It appears that the sign of the charge on the particle depends to some extent on the number of collisions made with the gas molecules and may be either positive or negative. Recoil phenomena probably play a part in the interpretation of some of the observed effects.

¹³ L. Wertenstein, *Compt. rend.*, 1909, **149**, 268; *A.*, ii, 713.

¹⁴ Mm. Curie, *ibid.*, 1908, **147**, 379; *A.*, 1908, ii, 797; H. Herschfinkel, *Le Radium*, 1909, **6**, 228.

¹⁵ W. Makower, *Le Radium*, 1909, **6**, 50.

¹⁶ S. Russ, *Phil. Mag.*, 1908, [vi], **15**, 601 and 737; *A.*, 1908, ii, 552, 556; W. T. Kennedy, *ibid.*, 1909, [vi], **18**, 744; *A.*, ii, 955.

The Uranium-Radium Disintegration Series.

Uranium Minerals.—A valuable set of measurements, giving the relative α -ray ionisation due to each member of the uranium-radium disintegration in equilibrium in radioactive minerals, in terms of that given by the parent substance, uranium, as unity, is embodied in the following table¹⁷:

Uranium.....	1.00	Radium- <i>B</i>	0.04 (?)
Ionium	0.34	Radium- <i>C</i>	0.91
Radium	0.45	Radium- <i>F</i>	0.46
Radium emanation	0.62	Actinium and products..	0.28
Radium- <i>A</i>	0.54		

Total activity..... 4.64 times uranium

Omitting radium-*B*, the activity of which is due to very soft β -rays, the actinium products, and uranium itself, the numbers are in good agreement, considering the ranges of the various α -particles expelled, with what is to be expected if each product gives one α -particle per atom disintegrating. But the results indicate clearly that uranium gives slightly more effect than would be the case if two α -particles were expelled. It is not impossible that in uranium two α -ray changes may succeed each other too rapidly for analysis. But as the range of the α -particles of uranium is low, this is not very probable. It is more likely, if two successive changes are really involved, that complete chemical similarity between the uranium and its product has hitherto prevented their separate identification (compare footnote, p. 262). Be that as it may, the result bears out in a most striking manner the atomic weight relations existing between uranium and radium, for the difference of 12 units indicates that three α -particles are expelled in the course of the transformation. Since ionium, the direct parent of radium, gives an α -particle, this requirement is entirely satisfied.

In the case of actinium and its products, for which the data are rather uncertain owing to the difficulties in completely separating it from the mineral, it is clear the result is quite exceptional. In the actinium series, four successive changes, possibly five, in which α -particles of average range are expelled, are known to occur. This result is the chief reason for considering actinium to be not in the main uranium-radium series, but in a branch or subsidiary series from uranium. The results would be consistent if out of about every eight atoms of uranium disintegrating seven produced radium and one actinium. The hypothesis has been put forward¹⁸

¹⁷ B. Boltwood, *Amer. J. Sci.*, 1808, [iv], 25, 269; *A.*, 1908, ii, 454; H. N. McCoy and W. H. Ross, *J. Amer. Chem. Soc.*, 1907, 29, 1698; *A.*, 1908, ii, 80.

¹⁸ H. N. McCoy and W. H. Ross, *ibid.*, 1698; *A.*, 1908, ii, 80; F. Soddy, *Phil. Mag.*, 1909, [vi], 18, 739; *A.*, ii, 952.

that the same atom may disintegrate in more than one way, each mode obeying the simple law independently of the other mode of disintegration. This would be extremely difficult to establish by experiment, for were one type of rays (say α -rays) given out in one mode of disintegration and another (say β -rays) in another, each radiation would decay with the *same* period, which would be the sum of two separate periods. The same fixed proportion would, however, exist between the amounts of products of the two modes of disintegration as existed between the reciprocals of the separate periods. Thus, if at one point in the disintegration series, one of the products disintegrated in dual fashion, the period of the one mode in which actinium was the product being about seven times the period of the other mode which gave rise to radium, the known relations between actinium, radium, and uranium would receive an easy explanation.

Doubt has been cast upon the complete constancy of ratio between the quantities of radium and uranium in minerals.¹⁹ This ratio has been redetermined by separating the two constituents chemically. For autunite a relatively smaller number (2.85), and for thorianite a relatively larger number (4.19), was obtained than the value for Joachimsthal pitchblende, which agreed with the accepted value (3.58×10^{-7}). It appears that the ratio is the higher the older the mineral, although the differences are not great. It would be interesting to have also measurements of the radium, for comparison, in the same minerals not subjected to any chemical processes of separation.

It is to be noted that the anomalous uranium-free radium mineral from Issy l'Eveque, the radium of which appears to have been derived from a distant uranium-lode by the action of percolating water, has its activity, which in the lump exceeds that of pure pitchblende, entirely confined to the surface. Acids dissolved off practically all the activity, but hardly any of the mineral itself.²⁰

As convenient standards of radioactivity have been suggested films of pure uranium oxide, U_3O_8 (about 0.7 gram, spread uniformly over a circular plate 7 cm. in diameter by means of chloroform), for which the α -activity is the maximum. The total α -activity of 1 gram of uranium, that is, the activity in a practically infinitely thin film, if no absorption occurred in the substance or its support, is equal to that of 796 sq. cm. of such a film. The saturation current per sq. cm. of surface of these films

¹⁹ Mlle. Gleditsch, *Compt. rend.*, 1909, **148**, 1451; **149**, 267; *Le Radium*, 1909, **6**, 165; *A.*, ii, 533, 714.

²⁰ H. N. McCoy and W. H. Ross, *J. Amer. Chem. Soc.*, 1907, **29**, 1698; *A.*, 1908, ii, 80.

is 5.79×10^{-13} ampere. They can thus be used to calibrate the readings of electroscopes in absolute measure. The α -activity of pure radium, free from products, is given as 1.3×10^6 times that of equal weight of uranium.²¹

The manufacturers' designation of the activity of radium preparations, as so many million times uranium, are meaningless and misleading. The best method is to compare the γ -activity of a known weight of the radium sample against a known weight of a standard preparation, under identical conditions, both preparations having been kept sealed up in glass tubes (provided with platinum wires sealed through) for at least a month. The purity of the primary standard should be checked by atomic weight estimations, or by the rate of evolution of heat. The ready spontaneous decomposition of radium bromide must be borne in mind.²² A specimen initially of 0.5 gram of pure radium bromide, as prepared by the maker, was found on delivery to weigh only 0.388 gram, and to be partly insoluble in water, but soluble in hydrobromic acid with evolution of carbon dioxide and gain of weight. Afterwards for some months the solution evolved bromine and relatively little oxygen. Then the evolution of bromine stopped, and the normal evolution of electrolytic gas began.

Radio-uranium.—The separation of a new substance, acting as the direct parent of uranium-*X*, and therefore intermediate between it and uranium, has been described.²³ In preparing uranium-*X* from a large quantity of uranyl nitrate, it was noticed that in one preparation the activity due to uranium-*X* had increased ten times on keeping. From this preparation uranium-*X* was separated, decaying at its normal rate, while the remainder generated a fresh amount of uranium-*X*. Very little of the substance so far has been separated, and it must resemble uranium in chemical nature very closely. The evidence, so far as it goes, is clear, but further confirmation must be awaited before the existence of the body can be regarded as established. There is no evidence whether or no it gives any specific radiation.

Uranium-X.—The repeated separation of uranium-*X* produced from a very large quantity of uranyl nitrate, in order to obtain evidence as to the nature of the product of its disintegration, has given results difficult to interpret on the view that this substance is intermediate between uranium and radium in the disintegration series. Were uranium-*X* the direct parent of ionium (which in

²¹ H. N. McCoy and G. C. Ashman, *Amer. J. Sci.*, 1908, [iv], **26**, 521; *A.*, ii, 148.

²² Sir W. Ramsay, *Sitzungsber. K. Akad. Wiss. Wien.*, 1908, **117**, (iia), 943; *Monatsh.*, 1908, **29**, 1013; *A.*, ii, 7.

²³ J. Danne, *Compt. rend.*, 1909, **148**, 337; *Le Radium*, 1909, **6**, 42; *A.*, ii, 288.

turn is the direct parent of radium and gives out α -rays during the transformation), it is to be expected that as its β -radiation decayed, a very feeble α -radiation, due to the ionium produced, would grow. By deviating the β -radiation as much as possible in a strong magnetic field and working in hydrogen, experiments have been made which would suffice to detect such a growth if it had occurred at the rate indicated by theory, and these experiments have given a negative result.²⁴ Although still incomplete, they raise the question whether uranium-*X* is not in the actinium rather than in the radium series.

The absorption of uranium-*X* by charcoal has shown that, for any one initial concentration of uranium-*X*, the ratio of the concentration in the solution and in the charcoal, when equilibrium is attained, is a constant independent of the mass of the charcoal employed.²⁵ This is analogous to Henry's Law for the solution of gases in liquids. A remarkable point seems to be that the absorption is dependent upon the presence of unknown impurities in the solution, and is completely prevented by a trace of thorium sulphate.²⁶

Parent of Radium.—The growth of radium in solutions of large quantities of purified uranyl nitrate has now been established to take place, and it is found to proceed at a rate proportional to the square of the time. This indicates that there can exist only one intermediate substance in the uranium-radium series, having a life-period which is long compared with the time of the experiments.²⁷ The period of average life of this substance, which is already known to be ionium, is calculated from these experiments to be 18,500 years as a maximum. If, however, there exist new intermediate substances in the series, of life-period comparable with the time of the experiments (four years), of which there is some evidence, the calculated period referred to would be reduced.²⁸

Intensely active preparations of ionium have now been prepared

²⁴ F. Soddy, *Phil. Mag.*, 1909, [vi], **18**, 858; *A.*, 1910, ii, 10.

²⁵ A. Ritzel, *Zeitsch. physikal. Chem.*, 1909, **67**, 725; *Le Radium*, 1909, **6**, 342; *A.*, ii, 851.

²⁶ Compare B. Szilárd, *Compt. rend.*, 1909, **149**, 113; *A.*, ii, 715.

²⁷ It may be remarked that the existence of a direct intermediate product so chemically alike to the parent that its full equilibrium quantity in the latter is never diminished by any chemical operation, which is of course always probable, is not considered in this and all similar deductions. The two successive substances would act always together exactly as one substance, and it is in this sense the deduction is to be read. On the other hand, if the two substances are not consecutive but are separated by an intermediate substance chemically dissimilar, as in the thorium series, the complete chemical similarity is no insuperable barrier to their separate identification.

²⁸ F. Soddy, *Phil. Mag.*, 1908, [vi], **16**, 632; *A.*, 1908, ii, 919; 1909, **18**, 846; *A.*, 1910, ii, 10.

both by the method of adding a little thorium to the mineral and separating out the thorium and ionium together,²⁹ and by precipitating the rare earths in the mineral with hydrofluoric acid in strongly acid solution.³⁰ It has also been prepared in the technical production of radium from pitchblende residues.³¹ The poverty of the latter in ionium led to the belief that it might be almost entirely separated, during manufacture, with the uranium, but the latter investigation shows that at least some is contained in the non-uranium residues. One estimate gives 20 per cent. of the full equilibrium amount. It has been established that ionium gives α -rays of range 2.8 cm. in air. But it does not give β -rays, previous observations to the contrary³² being due to the presence of uranium-X. Ionium possesses all the good qualities which have made polonium preparations of value in experimental work, with the added advantage that its activity is practically constant. The production of radium from these preparations has been proceeding at a constant rate since observations began, which is additional evidence of the extended period of life of ionium.

Radium.—Detailed accounts of the methods employed in the technical production of radium have been published, but the principles are identical with the original ones worked out by Debierne.³³ It has been shown that no radioactive matter is lost during the roasting, or in the liquors from the precipitations.³⁴ A new and very direct method for the determination of the period of average life of radium has been worked out,³⁵ depending on the complete separation of the full equilibrium amount of ionium present in a mineral and the measurement of the fraction of the equilibrium amount of radium it generates per year. The reciprocal of this fraction is the period of average life of radium. The accuracy of the method is independent of errors in the radium standards employed, and depends only on the completeness of the separation of all the ionium in the mineral. The value ascribed by this method to the period of average life was 2874 years, or for the period of half-change about 2000 years. This, which is necessarily, if anything, too high rather than too low,

²⁹ B. Boltwood, *Amer. J. Sci.*, 1908, [iv], **25**, 365; *A.*, 1908, ii, 455.

³⁰ B. Keetman, *Jahrb. Radioaktiv. Elektronik*, 1909, **6**, 265; *A.*, ii, 852; W. Markwald and B. Keetman, *Ber.*, 1908, **41**, 49; *A.*, 1908, ii, 144.

³¹ S. Meyer and E. von Schweidll-r, *Wiener Anzeiger. Sitzung.*, 11/6/09.

³² *Ann. Report*, 1907, 330.

³³ L. Haitinger und K. Ulrich, *Sitzungsber. K. Akad. Wiss. Wien*, 1908, **117**, (iia), 619; *Monatsh.*, 1908, **29**, 485; *A.*, 1908, ii, 857; H. Paweck, *Zeitsch. Elektrochem.*, 1908, **14**, 619; *A.*, 1908, ii, 917.

³⁴ J. Štěp, *Oesterr. Zeitsch. Berg. Hüttenwesen*, 1909, **57**, 155 and 173; *A.*, ii, 635.

³⁵ B. Boltwood, *Amer. J. Sci.*, 1908, [iv], **25**, 493; *A.*, 1908, ii, 551.

is about 12 per cent. greater than the value deduced from the method of counting the number of α -particles expelled.

The spectroscopic data, from which the high value of 257.8 had been assigned to the atomic weight of radium, have been held to be capable of a different interpretation. The earlier value is based on a relation which is shown not to be exact for the case of the known atomic weights of the other elements in the alkaline-earth family. By means of a new interpolation formula which takes into account these differences, the experimentally found value 226.5 has been deduced from the same data.^{35a} Obviously the spectroscopic evidence is of little weight.

Thorium.

Further work on the β -rays of mesothorium³⁶ has shown that the latter substance consists of two successive products, for which the names thorium-1 and thorium-2 are suggested, the next product, radio-thorium, being, with this nomenclature, thorium-3. Starting from meso-thorium freed from radio-thorium by repeated purification, and precipitating in the solution a little added zirconium by means of ammonia, the precipitate contains thorium-2, possessing a β -radiation which decays exponentially to zero in some hours, whilst the filtrate yields a substance (thorium-1) giving at first no β -rays, but regenerating them exponentially with the same period as that at which the β -radiation of the first preparation decays. The period of half-change is given as 6.2 hours. The series now runs:

The half-period of radio-thorium (737 days) is obtained by direct observation,³⁷ and from it and the curves of the variation of activity of freshly-made thorium preparations with time, that of meso-thorium (thorium-1) is deduced to be 5.5 years.³⁸ Of the total α -activity of thorium in equilibrium with all its products, it has been deduced very neatly that 11 per cent. is due to thorium

^{35a} W. Marshall Watts, *Phil. Mag.*, 1909, [vi], 18, 411; *A.*, ii, 780.

³⁶ *Ann. Report*, 1907, 326; O. Hahn, *Physikal. Zeitsch.*, 1908, 9, 245 and 246; *A.*, 1908, ii, 454.

³⁷ *Ann. Report*, 1907, 324.

³⁸ H. N. McCoy and W. H. Ross, *J. Amer. Chem. Soc.*, 1907, 29, 1709; *A.*, 1908, ii, 81; O. Hahn, *Physikal. Zeitsch.*, 1908, 9, 392; *A.*, 1909, ii, 557.

itself, 20 per cent. to radio-thorium, and 69 per cent. to thorium-*X* and later products. The total α -activity of 1 gram is 1009 times that of 1 sq. cm. of the standard film of uranium oxide already described.

Atmospheric and Natural Radioactivity.

The amount of radium emanation in the atmosphere has been determined by condensation in charcoal at the ordinary temperature,³⁹ by condensation at liquid air temperature in metal tubes,⁴⁰ and in other ways.⁴¹ The amount varies widely in different localities and for the same place with atmospheric conditions, and is of the order of the equilibrium amount from 10^{-11} to 10^{-10} gram of radium per cubic metre of air. It is insufficient to cause more than a small part of the natural ionisation in the air, which is ascribed mainly to the penetrating radiation from radioactive elements in the earth's surface.⁴² The radium emanation has been detected in the atmosphere by means of its active deposit at great heights, up to 3000 metres, by the use of free balloons.⁴³

A number of investigations have shown that the thorium emanation plays a large part in producing the active deposit from the atmosphere, and that thorium is an agent in terrestrial radio-activity of importance comparable with uranium. In Rome⁴⁴ and Manchester⁴⁵ the equilibrium quantity of active deposit on a negatively charged wire after long exposure to the atmosphere was found to be mainly due to thorium. In New Haven (Connecticut) and California⁴⁶ the proportion is rather less, while at high places in the Alps little thorium could be detected.⁴⁷ The proportion probably depends somewhat on the potential to which the wire is charged. For Manchester it has been calculated that 1.4 times more thorium than uranium atoms break up per second in the earth's crust, so that there must be seven times more thorium present than uranium. Direct methods for the determination of thorium in common rocks have been worked out.⁴⁸ The Vesuvian

³⁹ J. Satterly, *Phil. Mag.*, 1908, [vi], **16**, 584; *A.*, 1908, ii, 918; A. S. Eve, *ibid.*, 622; *A.*, 1908, ii, 919.

⁴⁰ G. C. Ashman, *Amer. J. Sci.*, 1908, [iv], **26**, 119; *A.*, 1908, ii, 918.

⁴¹ W. Wilson, *Phil. Mag.*, 1909, [vi], **17**, 216; *A.*, ii, 205.

⁴² K. Kurz, *Physikal. Zeitsch.*, 1909, **10**, 834; Th. Wulf, *ibid.*, 997.

⁴³ S. Flemming, *ibid.*, 1908, **9**, 801; *A.*, ii, 7.

⁴⁴ G. A. Blanc, *ibid.*, 295; *A.*, 1908, ii, 452.

⁴⁵ W. Wilson, *Phil. Mag.*, 1909, [vi], **17**, 321; *A.*, ii, 202.

⁴⁶ H. Dadourian, *Le Radium*, 1908, **5**, 102; *A.*, 1908, ii, 453; F. A. Harvey, *Physikal. Zeitsch.*, 1909, **10**, 46; *A.*, ii, 203.

⁴⁷ A. Gockel and Th. Wulf, *ibid.*, 1908, **9**, 907; *A.*, ii, 109.

⁴⁸ G. A. Blanc, *Le Radium*, 1909, **6**, 306; *Phil. Mag.*, 1909, [vi], **18**, 146; J. Joly, *ibid.*, 1909, [vi], **17**, 760; **18**, 140, *A.*, ii, 458, 637; R. J. Strutt, *Proc. Roy. Soc.*, 1909, **83**, A, 97; *A.*, 1910, ii, 9.

lavas appear unusually rich both in thorium and radium. Fifty-one rocks from the St. Gothard Tunnel show on the average about two-thirds as much thorium as uranium (calculated from the radium). In the soil of Rome 1.45×10^{-5} gram of thorium per gram was found, and larger quantities in syenites and granites.

As an agent in the production of natural radioactivity, and in questions connected with the heat of the interior and underground temperature gradients, the importance of thorium appears co-equal with that of the uranium-radium series. On the important question as to the amount of radium in the ocean, considerable discrepancies exist in the results of different workers, and discussion may be deferred.⁴⁹ The amount is extremely small, although in the aggregate it must total a very large quantity.

Reference may be made to a work where the main geological bearings of radioactivity have been fully discussed,⁵⁰ which renders it the less necessary to deal with them in detail here.

Potassium.

A number of investigations have left no doubt as to the existence of a specific feeble β -activity from salts of potassium, which in all probability is a specific property of the element.⁵¹ Rubidium has also been shown to possess a distinctive β -activity far less penetrating than that of potassium.⁵² The value of $\lambda/d(\text{cm.})^{-1}$ of the potassium β -ray is given as 8, showing that it is only a little less penetrating than the radiation of uranium-X. For the rubidium radiation the value is 53. The activity of very thick layers of potassium salts is slightly greater than that of rubidium, but the specific activity of the latter, allowing for its greater absorption, must be seven times as great as the former. All attempts to concentrate the activity seem to have failed.^{52a}

Various.

Three other notable results may be briefly mentioned. The speed of crystallisation of superfused droplets of pure sulphur,

⁴⁹ J. Joly, *Phil. Mag.*, 1908, [vi], **15**, 385; **16**, 190; *A.*, 1908, ii, 246, 649; 1909, **18**, 396; *A.*, ii, 780; A. S. Eve, *ibid.*, 102; *A.*, ii, 633; W. Knoche, *Physikal. Zeitsch.*, 1909, **10**, 157; *A.*, ii, 287.

⁵⁰ J. Joly, *Radioactivity and Geology*. Constable and Co., 1909.

⁵¹ J. C. McLennan and W. T. Kennedy, *Phil. Mag.*, 1908, [vi], **16**, 377; *A.*, 1908, ii, 750; M. Levin and R. Ruer, *Physikal. Zeitsch.*, 1908, **9**, 248; *A.*, 1908, ii, 448; W. W. Strong, *Physical Rev.*, 1909, **29**, 170; compare *A.*, ii, 715; E. Henriot, *Compt. rend.*, 1909, **148**, 910; *A.*, ii, 458; E. Henriot and G. Vavon, *ibid.*, **149**, 30; *A.*, ii, 635; E. H. Büchner, *Proc. K. Akad. Wetensch. Amsterdam*, 1909, **12**, 154; *A.*, ii, 779.

⁵² N. Campbell, *Proc. Camb. Phil. Soc.*, 1909, **15**, 11; *A.*, ii, 288.

^{52a} Compare, however, E. Ebler, *Chem. Zeit.*, 1908, **32**, 812.

under the microscope, has been found to be increased by the β -rays of radium, whereas X -rays produced no effect. The effect is possibly to be associated with the charge carried by the β -rays.⁵³ The pleochroic halo in certain minerals has been associated with the α -rays of embedded granules of radioactive minerals producing changes in colour in the surrounding matrix. The radius of these halos corresponds closely with the range of the α -particle in the mineral.⁵⁴ In this way visual evidence has been obtained of quantities of radioactive matter only just within the range of the most sensitive electrical methods. A sensitive micro-balance on a new principle has been constructed capable of detecting changes in the weight (of an otherwise small constant load) if greater than 4×10^{-9} gram, which it is proposed to attempt to employ in following the weight changes in radioactive matter due to disintegration.⁵⁵ The balance, in the first place, is a gravity balance of the ordinary type, entirely constructed of fused quartz, the beam weighing less than 0.5 gram, carrying on one end as counterpoise a sealed quartz bulb containing a known amount of air. Balance is effected by varying the external pressure of the air in the balance case. Accordingly, as the external pressure is less or greater than that of the air in the bulb, the latter represents a small positive or negative weight of easily determinable amount. The pressure of the air in the balance case is adjusted until the beam remains at zero position.

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⁵³ L. Frischauer, *Compt. rend.*, 1909, **148**, 1251; *Le Radium*, 1909, **6**, 161; *A.*, ii, 532.

⁵⁴ J. Joly, *Phil. Mag.*, 1907, [vi], **13**, 381; O. Mügge, *Centr. Min.*, 1907, 397; 1909, 65, 113, and 143; *A.*, ii, 286.

⁵⁵ B. D. Steele and K. Grant, *Proc. Roy. Soc.*, 1909, **82**, A, 580; *A.*, ii, 876.