



LXI. A comparison of the coefficients of diffusion of thorium and actinium emanations, with a note on their periods of transformation

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interfere. For instance, on the following dates, wet weather, unaccompanied by wind, is followed by a rise in the amounts of emanation at 50 cm.:—Sept. 4, 12, 23–29, Oct. 3, 14, 23, Nov. 8, Dec. 26, Jan. 5, 17–21, 25, Feb. 8, 18. The only marked exception occurs on Oct. 27–29.

An examination of the barometric chart for the period covered by the experiments does not reveal any influence of atmospheric pressure on the results, except in so far as its fluctuations are accompanied by storm, rain, &c.

The average amounts of emanation found in these experiments should be of interest. They are as follows:—

Ground-gas (102 experiments) 163×10^{-12} curie per litre.

Exhalation (98 experiments) 2673×10^{-12} curie per sq.m. per hour.

In the course of the previous experiments referred to*, 38 observations on *ground-gas* were made, at the same place as the present ones, but at depths varying from 20 to 150 cm. The average of these was 180×10^{-12} curie per litre, which agrees fairly well with that given above.

The previous observations on *exhalation* were not made at the same spot. Ten of them were made about 30 ft. from the present ones, and 3 or 4 ft. from the wall of the laboratory, and twelve were made at a spot three miles away. These two sets gave averages of 839×10^{-12} and 4087×10^{-12} respectively.

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Trinity College, Dublin,
July 1, 1912.

LXI. *A Comparison of the Coefficients of Diffusion of Thorium and Actinium Emanations, with a Note on their Periods of Transformation.* By MAY SYBIL LESLIE, M.Sc.†

THE values hitherto found for the diffusion coefficients of radium and thorium emanations differ considerably from one another. They are in general, however, except in the case of Perkins's experiments with radium emanation‡, higher than would be expected from the probable molecular weights calculated on the basis of the disintegration theory, and practically confirmed by the values of the

* *Loc. cit.*

† Communicated by Prof. E. Rutherford, F.R.S.

‡ P. B. Perkins, Amer. Journ. of Sci., June 1908 xxv. p. 461.

effusion coefficients found by Debiérne* for radium emanation and by the author † for thorium emanation. No other data than the diffusion coefficient of the emanation are available for determining the molecular weights in the actinium series, and the results obtained in this direction suffer from the same uncertainty as in the case of the other emanations. For example, Debiérne ‡ and Bruhat § both obtained .112 as the diffusion coefficient in air at 76 cm. pressure and 15° C., whereas Russ || obtained .123. One would expect, however, to find the same disturbing cause, whatever it may be, operating in the diffusion experiments for all the radioactive gases. Therefore a comparison of the rates of diffusion of thorium and actinium emanations under strictly identical conditions should give the ratio of their molecular weights. If the value for thorium emanation is taken as 220, that for actinium emanation may then be calculated.

Such a comparison was the object of the present investigation. Unfortunately, through the discovery of an unexpected source of error, no very definite conclusions could be drawn from the results, but as these throw some light on the cause of the discrepancies between the observed values for the diffusion coefficients, it will be useful to record them.

Experimental Arrangements.—The diffusion vessel was a glass cylinder of length 50 cm. and internal diameter 5 cm. It was closed at the top by a hollow ground-glass stopper to which was sealed, for communication with the exterior, a tube with two branches each provided with a tap. The bottom of the vessel was covered by a tray over which the substance providing the emanation was uniformly spread. The vessel was surrounded by a water-jacket, which kept it at a temperature sufficiently constant for the requirements of the experiment.

Preliminary Experiments with Thorium Emanation.—Thorium hydroxide dried at a gentle heat was used as the emanating source, and the distribution of emanation was at first measured by the active deposit on a metal plate down the centre of the tube. To both sides of the plate, across the end near to the substance, strips of tinfoil of equal width, about 4 mm., were attached. The exposures were from 15

* A. Debiérne, *Comptes Rendus*, juin 1910, p. 1740.

† M. S. Leslie, *Comptes Rendus*, juillet 1911, p. 328.

‡ A. Debiérne, *Le Radium*, 1907, p. 213.

§ Bruhat, *Le Radium*, 1909, p. 87.

|| S. Russ, *Phil. Mag.* xvii. p. 412 (1909).

to 20 hours, and no electric field was applied. Four or five hours after the removal of the plate, when the active deposit was decaying according to an exponential law, and corrections could be more conveniently applied, the activities of the strips in pairs, one from each side at the same distance from the bottom, were measured in a sensitive α -ray electro-scope.

Within the limits of experimental error the emanation ought to be distributed according to an exponential law*

$p = p_0 e^{-\sqrt{\frac{\lambda}{D}}x}$, where p_0 is the initial concentration, p the concentration at a point at distance x , λ the radioactive constant, and D the coefficient of diffusion. If the active deposit reaches the plate at the level at which it is formed, the variation in the activity of the strips should follow the same law. This was not the case, however. The logarithm of the activities plotted against the distances x did not give a straight line but a curve, according to which the coefficient of diffusion apparently increased with the distance from the source.

Two parallel plates, 2 or 3 mm. apart, without a field, as in Debierne's and Bruhat's experiments with actinium emanation †, were then substituted, the distribution of emanation between them being examined; but still departures from an exponential law were observed. It was thought probable that this might be caused by the diffusion of some of the active deposit so that it reached the plates at a level other than that at which it was formed from the emanation. According to some observations made by Mme. Curie on radium deposit ‡, if the air containing emanation is not perfectly dry, agglomerations of molecules of water vapour and active deposit are probably formed. As the phenomenon observed might have been due to these, a tube of phosphorus pentoxide was placed in the cylinder to dry the air, but apparently a certain amount of disturbance still took place.

Next an electric field was employed § to direct the active deposit more rapidly to the plates. With 2-4 volts, and even with 40 volts, some trace of the same phenomenon remained; but with 80 volts practically a straight line was obtained when the logarithms of the activities were plotted against distance. In all subsequent experiments a difference

* Rutherford, 'Radioactivity,' p. 276.

† *Loc. cit.*

‡ Mme. Curie, *Comptes Rendus*, 1907, pp. 477 & 1145.

§ Rutherford, *loc. cit.*; Russ, *loc. cit.*

of potential of 160 volts was used. The measurements were begun about 1.5 cm. from the end of the cathode plate, and the strips of tinfoil did not extend the whole width of the plate, but were several millimetres shorter ; so that any effects due to non-uniformity of the field at the edge of the plate might be avoided. Measurements were made at atmospheric pressure, and also at reduced pressures. The product of the pressure P and the coefficient of diffusion D should be a constant at constant temperature, varying as the square of the absolute temperature. The values of $P \times D$ given below have been corrected for 15°C . The coefficient of diffusion was calculated assuming 54 seconds * as the period of transformation of thorium emanation, or $\lambda = 1.29 \times 10^{-2} \text{ sec.}^{-1}$.

P.	$P \times D$ (corrected).
76.2 cm.	6.31
76.7 "	6.42
75.8 "	6.07
75.65 "	6.69
75.5 "	6.00
73.8 "	6.43
54.95 "	6.74
37.2 "	6.80
31.2 "	6.89
29.7 "	6.44
12.8 "	6.20
8.8 "	6.39
	Mean .. <u>6.46</u>

The value of D at 76 cm. pressure and 15°C . = 0.85.

The apparatus was sufficiently long to prevent the necessity for additional corrections at reduced pressures.

Experiments with Actinium — The thorium hydroxide was replaced by an actinium preparation, and the rate of diffusion determined in exactly the same way. All the experiments were made at reduced pressures, since at atmospheric pressure only a very small quantity of emanation reached the point at which the measurements were begun. An ordinary α -ray electroscope was used instead of the sensitive one employed in the experiments with thorium emanation. The period of transformation of actinium emanation † was taken as 3.9 secs. or $\lambda = 1.8 \times 10^{-1} \text{ sec.}^{-1}$.

* See note later.

† See note at the end.

P.	P × D (corrected).
42.75 cm.	7.50
42.0 "	8.30
36.95 "	6.80
33.25 "	7.40
32.5 "	6.77
20.7 "	7.23
19.9 "	7.50
18.2 "	6.82
17.45 "	7.87
15.9 "	7.25
15.7 "	7.44
	Mean .. 7.44

The value of D in air at 76 cm. pressure and 15° C. = .098.

Experiments with Glass Plates.—The values of the product P × D for actinium show even larger variations than those for thorium emanation. It was thought that perhaps the brass plates, about .5 mm. thick, were not sufficiently rigid and that they did not always occupy the same relative positions. The measurements for actinium were therefore repeated, using glass plates 2 mm. thick, covered with tinfoil on the sides facing one another, except at the edges, which were left uncovered for insulating purposes.

P.	P × D (corrected).
43.75 cm.	8.57
39.8 "	8.17
34.35 "	8.04
32.55 "	8.23
27.25 "	7.70
25.0 "	8.07
22.2 "	7.94
17.6 "	7.95
14.35 "	8.41
11.85 "	7.87
	Mean .. 8.12

The value of D in air at 76 cm. pressure and 15° C. = .107.

Evidently the form of the apparatus has a considerable effect on the results.

If the value of P × D for thorium emanation is altered in the same degree, it should now be about 7.13. To test this point the determinations with thorium emanation were

repeated, but with the difference that a small quantity of a strong radiothorium product was used instead of thorium hydroxide. Since the activities were not determined for at least five hours after the removal of the plates from the diffusion vessel, no difficulties would arise owing to the presence of radium emanation, because any radium active deposit would then have disappeared.

In the first experiments made the activities were eighty or ninety times those obtained in the former experiments, and the resulting value for $P \times D$ was found to be very high.

P.	$P \times D$ (corrected).
77.1 cm.	8.63
77.1 "	10.48
76.6 "	9.34
76.6 "	8.70
76.1 "	9.19
76.0 "	8.29
75.8 "	8.88
Mean of last four ..	<u>8.77</u>

The value of D in air at 76 cm. pressure and $15^\circ \text{C.} = \underline{.115}$.

The supply of emanation was cut down by covering the radiothorium with perforated tinfoil and layers of blotting-paper. The activities were now diminished to about twenty times those obtained from the hydroxide, and it was found that the value of $P \times D$ had also decreased.

P.	$P \times D$ (corrected).
76.7 cm.	8.76
76.0 "	8.15
76.0 "	8.48
75.9 "	8.21
75.9 "	8.17
75.8 "	9.38
Mean ..	<u>8.51</u>

The value of D in air at 76.0 cm. pressure and $15^\circ \text{C.} = \underline{.112}$.

A further diminution of emanation until it was only about five or six times that obtained from thorium hydroxide was again followed by a decrease in the value of the product $P \times D$, but only to a slight extent.

P.	P × D (corrected).
76·7 cm.	8·58
76·5 „	8·42
76·3 „	8·54
76·0 „	8·58
76·0 „	8·92
75·8 „	8·49
75·5 „	8·36
75·0 „	8·11
75·0 „	8·07
42·0 „	9·02
26·7 „	8·06
16·0 „	8·05
	Mean . . 8·43

The value of D in air at 76 cm. pressure and 15° C = ·111.

Discussion of Results.—The results given are sufficient to show that apparently slight alterations in the form of the apparatus may produce considerable variations in the value of the diffusion coefficient as determined experimentally. The minor departures from constancy of the value P × D in experiments made with exactly the same apparatus and amount of emanation are probably due to slight deviations from parallelism of the plates. That other experimenters have found as great variations can be seen from the paper by Bruhat. He also gives a full list of his results, and deviations from the mean value to the extent of 14 or 15 per cent. may be noticed.

The experiments show further that even without any change in the apparatus it is possible to vary the value of D by varying the amount of emanation in the vessel. These two facts seem to indicate the reason, not only why, in the past, different experimenters have found different values for D, but also why there is a discrepancy between the observed and the calculated values. The disturbing cause is very probably the active deposit, a portion of which perhaps loses its charge by recombination with the ions in the air, and then behaves more or less as a gas, until it accidentally strikes one of the plates. The effect was naturally more pronounced where no field was employed, or only a single plate. Even when a field was employed the anode showed a small quantity of activity, about 5 or 6 per cent. of that on the cathode. This might have been due either to electrically neutral, or negatively charged particles. The portions of the active deposit producing the largest effect would be the

long-period substances thorium B or actinium B. Indeed, ultimately it was only the amounts of thorium B and actinium B which were found, since, when the measurements were made, sufficient time had passed since the removal of the plates to allow all other products brought from the diffusion vessel to disappear. Wellisch and Bronson*, in a recent paper on the distribution of the deposit of radium in an electric field, have shown that with a difference of potential of 160 volts not more than 80 per cent. radium deposit reaches the cathode, and even with 4000 volts only 91 per cent. was obtained. This, it is true, was across a distance of 28 mm. and not 2 mm. as in the present experiments.

With an increase in the amount of emanation there will be an increase in the ionization. Hence, more reaction between the ions and the active deposits resulting in a greater value of D may reasonably be expected. This would account for the high values obtained with radiothorium as source of emanation. Wellisch and Bronson, in the paper just referred to, state that only with fields of a few volts does any change in the amount of deposit collected appear to follow a change in the degree of ionization. One might assume, however, that a neutral particle could collide with an ion and become charged again. In the experiments referred to, the final result would be the same as if the particle had remained charged all the time. This would not be the case, however, in the diffusion experiments, where it is important that the active deposit should attach itself to the plate at the level at which it is formed.

Evidently, it is exceedingly difficult to say when the thorium and actinium emanations may be justly regarded as comparable with one another. This may perhaps have been approximately the case when amounts of emanation which were equally active were employed, that is when the value of D for thorium emanation was $\cdot 111$, and for actinium emanation $\cdot 107$. If such an assumption were made, the results would indicate that the molecular weights are very close to one another. More reliance, however, could be placed on this conclusion if the periods of all the thorium products bore the same ratio to one another as those of the actinium products. The diffusion of the active deposits might then be expected to produce proportionately the same effect in each case.

With regard to the absolute value of the coefficient of diffusion, it may be concluded that the smaller values found are more nearly correct than the larger ones. The smallest

* E. M. Wellisch and H. L. Bronson, *Phil. Mag.* May 1912 p. 714.

value was found for thorium emanation when only a very small amount of emanation was used, and when the plates were thin, *i. e.* metal plates .5 mm. thick instead of the glass plates 2 mm. thick employed later. What is the exact effect produced by the thickness of the plate it would be difficult to say, but it seems reasonable to suppose that a thin plate in the diffusion vessel would produce less disturbance than a thick one. The value obtained under these circumstances was .085. Therefore, one may say that at 76 cm. pressure and 15° C. the coefficient of diffusion of thorium emanation is not greater than, and may possibly be less than .085. Similarly the upper limit for the coefficient of diffusion of actinium emanation is .098.

Summary.

(1) The values found for the diffusion coefficients of the radioactive gases depend on the amount of ionization in the diffusion vessel, and on the arrangements made for collecting the active deposit.

(2) A comparison of the diffusion coefficients of thorium and actinium emanations under conditions as nearly identical as the differences of period of the different substances concerned will allow, indicates that the molecular weights of the two gases are probably very nearly equal.

(3) The real coefficients of diffusion of thorium and actinium emanations are not greater than, and probably less, than .085 and .098 respectively.

Note on the Periods of Transformation of Actinium and Thorium Emanations.

As a knowledge of the radioactive constants of the emanation is necessary for the calculation of the above results, it was thought desirable to employ values obtained by exactly similar methods. The decay of the emanations was followed directly by means of an electroscope.

In the first attempts made an emanation electroscope was partially evacuated, the leaf charged too much to be seen on the scale, and air containing a small amount of actinium emanation—not sufficient to drive the leaf entirely off the scale—was allowed to enter abruptly. After two or three seconds, when the leaf had appeared in the microscope, scale-readings were taken. The intervals of time were measured by means of a swinging pendulum placed near the electroscope, so that it could be watched at the same time as the scale was read. When the emanation had disappeared a

continuation of the readings gave the leak. The activity due to the active deposit formed in the 25 or 30 seconds elapsing since the introduction of the emanation was practically zero. After corrections had been made for the leak and for variations in the sensitiveness along different parts of the scale, the difference between the final reading and the reading at any given moment would be proportional to the total amount of emanation present at that moment. The logarithms of these values were plotted against time. In nineteen experiments the values for the period varied between 3.70 and 4.00 seconds, giving a mean of 3.86 seconds.

In the hope of attaining a greater degree of accuracy a modification of the above method was devised. The pendulum was insulated and kept charged to a constant voltage, and as in swinging it passed its lowest point, it touched the exposed tip of a fine insulated wire passing from the gold-leaf support to the outside of the electroscope. Thus at definite intervals of time (about 2 seconds) the leaf could be re-charged, its fall representing the average activity of the emanation during that time. When no emanation was present, there was always a certain amount of oscillatory movement of the leaf, due partly to induction effects produced by the charged pendulum on the electroscope, and partly to the leak. Correction was made for this. In twelve experiments, of which five gave 3.95 seconds, the variation was between 3.90 and 3.98 seconds, the mean being 3.95 seconds.

A further change was then introduced. The outside of the electroscope was charged and the pendulum earthed. An earthed metal plate screened the electroscope from the pendulum except at the small hole through which passed the wire in connexion with the leaf. In this way all disturbance due to induced charges was prevented. Sixteen experiments gave values varying between 3.81 and 4.06 seconds, the mean being 3.92 seconds.

The period of thorium emanation was also determined by this method. Of twelve experiments eleven gave values varying between 53.5 and 55.1 seconds, mean 54.3 seconds: the twelfth, 55.8 seconds, varying considerably from the others, was neglected.

As the variations of the mean values from the period of 3.9 seconds found by Debiere * by the air-current method for actinium emanation, were much less than the variations between the actual experimental values obtained above, this value, 3.9 seconds or $\lambda = 1.8 \times 10^{-1} \text{ sec.}^{-1}$ was retained in the

* Debiere, *Comptes Rendus*, 1904, p. 411.

calculation of the diffusion coefficient. Hahn and Sackur* obtained the same value by following the change of activity directly by means of an electrometer. Similarly the period of transformation, 54 seconds or $\lambda = 1.29 \times 10^{-2}$ sec.⁻¹ found by Bronson †, was adopted for thorium emanation.

My thanks are due to Professor Rutherford for his help and interest in these experiments.

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LXII. *The Ranges of the α particles from the Thorium and Actinium Products.* By H. GEIGER, Ph.D., Lecturer in Physics, and J. M. NUTTALL, M.Sc., University of Manchester ‡.

IN two previous papers § we have shown that there exists a quantitative relation between the range of an α particle and the transformation constant of the product from which it arises. For the products of the uranium-radium series the values of the ranges of the α particles were partly taken from papers by Bragg ||, and were in part redetermined. As regards the members of the actinium and thorium series, with a few exceptions, only the initial measurements of Hahn ¶ were available.

A determination of the ranges of the α particles from most of the products of these families is difficult, since, owing to the short periods of some of them, it is impossible to separate the products from each other. Thus the actinium emanation and actinium A will always be associated with actinium X, and therefore complicate a determination of the range of the α particles from the latter substance. Similar difficulties arise in the thorium series.

In the following we have undertaken the redetermination of the ranges of the α particles emitted by the products of the thorium and actinium series, making use of the experimental arrangement we have previously employed for the determination of the ranges of the α particles from uranium.

* Hahn and Sackur, *Ber. deutsch. Chem. Ges.* 38. ii. p. 1943 (1905).

† Bronson, *Amer. Journ. of Sc.* xix. p. 185 (1905).

‡ Communicated by Prof. E. Rutherford, F.R.S.

§ H. Geiger and J. M. Nuttall, *Phil. Mag.* xxii. p. 613 (1911); xxiii. p. 439 (1912).

|| W. H. Bragg and R. D. Kleeman, *Phil. Mag.* viii. p. 726 (1904); x. p. 318 (1905).

¶ O. Hahn, *Phil. Mag.* xii. p. 244 (1906).