

XVII. The relation between uranium and radium.—Part VI. The life-period of ionium

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XVII. *The Relation between Uranium and Radium.*—Part VI.
The Life-period of Ionium. By FREDERICK SODDY, M.A.,
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AN experimental examination of the question whether radium is produced from uranium has been in progress by one of us since 1902. A clear growth of radium in a uranium solution, initially purified from radium by precipitating barium sulphate in the solution, was observed in 1904 †, but the extreme slowness of the growth suggested the existence of a long-lived intermediate parent of radium, which was separated by Boltwood in 1907 and named ionium. The present series of experiments were started in conjunction with Mr. T. D. Mackenzie in 1905. Uranium preparations were purified as carefully as possible by methods designed to eliminate all other substances, so that neither radium nor the hypothetical intermediate parent of radium would initially be present ‡. Accounts of the progress of the measurements on the quantities of radium in the various solutions have been published from time to time §. In 1912,

* Communicated by the Authors.

† F. Soddy, 'Nature,' May 12, 1904; Jan. 26, 1905; Phil. Mag. [6] ix. p. 768 (1905); compare W. C. D. Whetham, 'Nature,' May 5, 1904; Feb. 2, 1905.

‡ F. Soddy and T. D. Mackenzie, Phil. Mag. [6] xiv. p. 272 (1907).

§ F. Soddy, Phil. Mag. [6] xvi. p. 632 (1908); xviii. p. 846 (1909); xx. p. 340 (1910).

Phil. Mag. S. 6. Vol. 30. No. 176. Aug. 1915. P

a connected account of the whole work up to that date was given in a lecture at the Royal Institution (March 15th, 1912)*, and the conclusions then drawn may be briefly restated.

In the following table particulars are given of the four uranium preparations studied :—

No.	Grams of Uranium.	Date of purification.	Method of purification.	Initial rate of growth of radium per year per kilogram of uranium.
I. ...	255	24/10/05	Ether.	13×10^{-12} g.
II. ...	278	14/ 8/06	Ether.	8×10^{-12} g.
III. ...	408	13/12/06	Ether.	3.5×10^{-12} g.
IV. ...	3000	4/ 6/09	Recrystallization.	2.5×10^{-12} g.

The methods of purification of the uranium adopted, extraction with ether in the case of the first three preparations and repeated recrystallization from water in the case of the fourth, are those generally employed to remove uranium X from uranium, and, since uranium X is now known to be isotopic with ionium, the best possible methods for removing ionium had unknowingly been employed. A very slow rate of growth was apparent in all four preparations, diminishing in order from 13×10^{-12} gram of radium per year per kilogram of uranium in the case of the first, to about one-fifth of this rate in the case of the fourth. These differences can only be due to the more successful elimination of initial ionium in the successive preparations, and prove that, in the first two preparations at least, the growth of radium is to be ascribed mainly to initial ionium. As Rutherford has pointed out, the growth of radium from uranium, if ionium is the only long-lived intermediate product, must proceed initially according to the square of the time. But in 1912 there was no evidence that the growth of radium in any of the preparations was proceeding other than linearly with the time. That is to say, it was certainly due, for the most part, at least in the case of the first preparations, to initial ionium that had survived the purification processes, and there was in 1912 no positive evidence that uranium was producing radium at all. On the assumption that the whole of the radium came from the uranium, the *minimum* possible period of average life of ionium can be found. This was deduced

* Trans. Royal Institution, 1912.

to be 80,000 years from the results with the third preparation, and 70,000 years from the results with the fourth. Recalculating on the same data as are used later in this paper, these periods must be increased by the factor 1.2, that is to 96,000 and 84,000 years respectively. If part of the growth of radium is derived from initial ionium the period of ionium, naturally, is increased. Since it was thought that even in these most highly purified preparations part of the growth at least must be due to initial ionium, the period of the latter was estimated as probably at least 100,000 years (now 120,000 years). In 1912, however, the direct experiments only fixed a lower limit to the value of the period, and gave no indication of the true period.

Measurements on the rate of growth of radium in the various preparations were continued till September 1914, in Glasgow, under the same conditions and with the same instrument as previously*, and indicated a clear increase of the rate of growth in the fourth and largest uranium preparation, containing 3 kilograms of uranium (element). This increased rate has been confirmed by subsequent measurements with a new instrument and under slightly altered conditions of measurement in Aberdeen, whither the preparations were all successfully transported. It is now possible to say that a definite growth of ionium from uranium has been experimentally observed, and to fix the true period of ionium approximately.

Method of Measurement.

Until the summer of 1914, the method of measurement was the same as previously adopted and described*. In October 1914, the whole of the preparations were successfully transported in their sealed flasks to Aberdeen, and a new electroscope had to be set up for the measurements. Advantage was taken of this necessary break in the continuity of the measurements to modify the method slightly to render the observations less time-consuming. Hitherto, all the measurements had been made with the leaf charged negatively, maintaining the leaf charged as it leaked away during the 3-hour interval between admission of the emanation and measurement, to avoid errors through changes in the distribution of the active deposit†. It is more convenient to charge the leaf positively, though the sensitiveness of the electroscope is thereby reduced, as then the instrument can

* *Phil. Mag.* [6] xviii. p. 847 (1909).

† *Ibid.* p. 850.

be kept discharged during the waiting interval without changing its constant. This is a point of considerable importance in measurements of radium by the emanation method, and had long ago been adopted in all other measurements by this method; but, in order to preserve continuity, the old method had been retained with all the measurements by the old instrument in this set of experiments. With the new electroscope the new plan was adopted. Very great care was taken to avoid any introduction of error by this change, and measurements taken with the same standards by the old and new magnifying-power methods agreed perfectly. In addition, the magnifying power of the new microscope was considerably less than that of the old, and the two changes together caused a reduction of the sensitiveness of the new instrument to about one-third of that of the old. The accuracy of the measurements, however, was not affected by these changes.

In former papers, somewhat different values have been employed for the ratio of radium to uranium in pitchblende, and all the former results have been recalculated to the same value, viz. 3.4×10^{-7} g. radium per gram of uranium. In the present work, a large number of new standards from carefully analysed uranium minerals were prepared, as it was found that the old standards, prepared in 1909, no longer agreed among themselves. As is well known, it is practically impossible to keep such standards indefinitely, owing to the tendency of part of the infinitesimal amount of radium present to precipitate out of solution.

In the following three tables are given particulars of the calibrations of the instruments. The first refers to the original instrument, the second to the new instrument, and the third to the latter after an accident to the gold leaf. The uranium minerals used were those employed in an earlier research*, on the ratio of radium to uranium in minerals. The radium-barium chloride preparations used were some containing about 10 mg. of radium (element) per kilogram, in which the radium has been determined by γ -ray measurements of spherical samples against a radium standard, according to the method described in 'Chemistry of the Radio-Elements,' Part I. Second edition, 1915, p. 93, in which the absorption of the γ -rays in the preparation itself is corrected for. Known weights of these were dissolved, and the solutions diluted to convenient strength. The electroscope was thus calibrated independently on the γ -ray standard

* F. Soddy and Miss R. Pirret, *Phil. Mag.* [6] **xx**. p. 345 (1910); **xxi**. p. 652 (1911).

and the ratio of radium to uranium required to make the results agree with those from uranium minerals was 3.4×10^{-7} .

Calibration of Electroscope.

I. Old Instrument.

Standard.	Uranium (mgs.).	Radium (grms. $\times 10^{-10}$)	Leak (divs. per minute).	Constant (grms. of Ra $\times 10^{-12}$ required to give 1 div. per min.).
Joachimstahl pitchblende ... (new standard)	0.4397	1.495	28.03	5.33
Joachimstahl pitchblende ... (old standard J. P. B.)	0.3539	1.203	22.40	5.37

Mean ... 5.35

II. New Instrument.

Standard.	Uranium (mgs.).	Radium (grms. $\times 10^{-10}$)	Leak (divs. per minute).	Constant.
Joachimstahl pitchblende 1 .	0.3366	1.141	7.81	14.65
Joachimstahl pitchblende 2 .	0.5388	1.832	12.60	14.53
Joachimstahl pitchblende 3 .	0.5164	1.756	12.26	14.32
Cornish pitchblende 1	0.3621	1.231	8.59	14.33
Cornish pitchblende 2	0.3620	1.231	8.59	14.33
Radium-barium chloride 1	13.487	92.55	14.57
Radium-barium chloride 2	13.463	94.84	14.19
Radium-barium carbonate 1	3.993	27.51	14.51
Radium-barium carbonate 2	3.544	24.06	14.73

Mean ... 14.46

On April 23, 1915, the tip of the leaf of the electroscope was broken off through an accident. The instrument with its shortened leaf was recalibrated.

Standard.	Uranium (mgs.).	Radium (grms. $\times 10^{-10}$)	Leak (divs. per minute).	Constant.
Joachimstahl pitchblende 1 .	0.3366	1.144	7.22	15.85
Joachimstahl pitchblende 2 .	0.5388	1.832	11.65	15.72
Radium-barium carbonate 1	3.993	25.10	15.90
Cornish pitchblende 2	0.3620	1.2310	8.04	15.30
Radium-barium chloride 2	13.463	86.68	15.53

Mean ... 15.66

Results.

The results of the measurements of the quantities of radium in the four preparations from the start are shown in the following tables.

Preparation No. I.

255 grms. Uranium. Purified October 24, 1905.

Date.	Time from start (years).	Leak (divs. per minute).	Constant (grms. Ra $\times 10^{-12}$ required to give 1 div. per min.).	Radium (grms. $\times 10^{-12}$).
Mean of first 9* tests	0 to 0.85	1.3	12	16
June 9, 1908.....	2.62	3.2	5.78	19
Aug. 8, 1908.....	2.78	3.17		18
Sept. 25, 1908.....	2.95	3.39		20
Nov. 20, 1908.....	3.07	3.44		20
May 5, 1909.....	3.53	3.92		23
June 14, 1909.....	3.64	4.2		24
July 27, 1909.....	3.75	4.0		23
Aug. 27, 1909.....	3.84	4.28		25
Sept. 29, 1909.....	3.93	4.16		24
Oct. 12, 1909.....	3.96	4.4	25	
June 3, 1910.....	4.61	4.7	5.2	24
Aug. 19, 1911.....	5.82	6.5	5.25	34
Jan. 20, 1912.....	6.24	6.2	5.3	33
July 30, 1914.....	8.76	7.33	5.35	39
Oct. 21, 1914.....	8.99	2.62	14.46	38
Nov. 26, 1914.....	9.09	2.55		37
May 5, 1915.....	9.51	2.21		15.66

* Phil. Mag. [6] xviii. p. 854 (1909).

Preparation No. II.

278 grms. Uranium. Purified August 14, 1906.

Date.	Time from start (years).	Leak (divs. per minute).	Constant (grms. Ra $\times 10^{-12}$ required to give 1 div. per min.).	Radium (grms. $\times 10^{-12}$).	
May 30, 1908.....	1.80	3.02	5.78	18	
May 7, 1909.....	2.73	3.68		21	
June 25, 1909.....	2.86	3.72		21	
Oct. 4, 1909.....	3.14	4.0		23	
June 4, 1910.....	3.80	4.28		5.2	22
Aug. 23, 1911.....	5.03	4.80		5.25	25
Jan. 30, 1912.....	5.46	5.12		5.3	27
July 29, 1914.....	7.96	6.13		5.35	33
Oct. 22, 1914.....	8.18	2.18		14.46	32
Nov. 27, 1914.....	8.29	2.20	32		
May 7, 1915.....	8.70	1.84	15.66		29

Preparation No. III.

408 grms. Uranium. Purified December 13, 1906.

Date.	Time from start (years).	Leak (divs. per minute).	Constant (grms. Ra $\times 10^{-12}$ required to give 1 div. per min.).	Radium (grms. $\times 10^{-12}$).
Aug. 8, 1908	1.66	0.67	5.78	4
Nov. 11, 1908 ...	1.93	0.77		4
May 6, 1909	2.39	1.25		7
June 15, 1909 ...	2.50	1.26		7
Oct. 1, 1909	2.80	1.37		8
March 4, 1910 ...	3.22	1.78	5.2	9
June 10, 1910 ...	3.49	1.6		8
Aug. 20, 1911 ...	4.68	3.5	5.25	18
March 8, 1912 ...	5.23	2.44	5.3	13
July 28, 1914 ...	7.63	4.08	5.35	22
Oct. 24, 1914 ...	7.86	1.50	14.46	22
Nov. 30, 1914 ...	7.96	1.51		22
March 16, 1915...	8.26	1.58		23
May 3, 1915	8.39	1.41	15.66	22

Preparation No. IV.

3000 grms. Uranium. Purified June 4, 1909.

Date.	Time from start (years).	Leak (divs. per minute).	Constant (grms. Ra $\times 10^{-12}$ required to give 1 div. per min.).	Radium (grms. $\times 10^{-12}$).
Aug. 26, 1909 ...	0.22	7.0	5.78	40
Sept. 28, 1909 ...	0.31	7.1		41
Dec. 9, 1909	0.51	7.9		46
March 3, 1910 ...	0.74	8.2	5.2	43
June 12, 1910 ...	1.02	8.3		43
Aug. 24, 1911 ...	2.21	10.6	5.25	56
Feb. 2, 1912	2.66	10.6	5.3	56
July 31, 1914 ...	5.15	19.4	5.35	104
Dec. 2, 1914	5.49	7.2	14.46	104
March 17, 1915...	5.78	8.1		117
May 4, 1915	5.91	7.4	15.66	116

These results are plotted in fig. 1, with time in years as abscissæ and quantities of radium ($\times 10^{-12}$ g.) as ordinates. In the case of Preparation II., 20 must be subtracted from the ordinates in the figure.

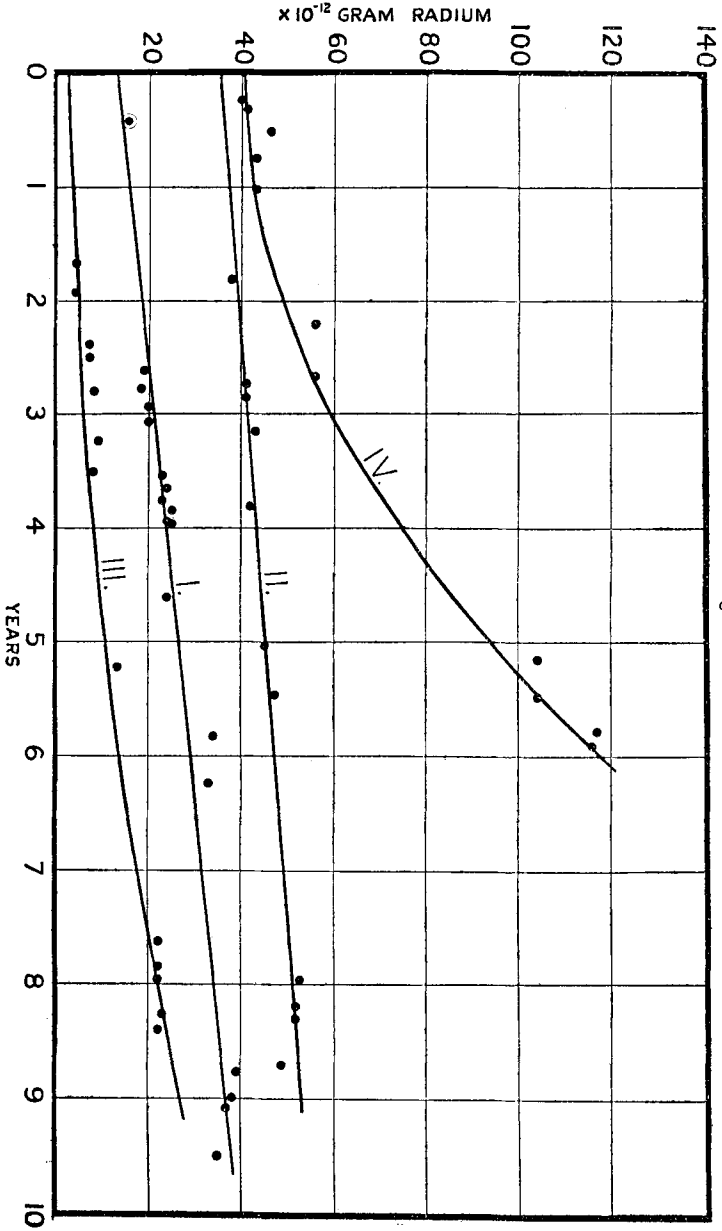


Fig. 1.

Discussion of Results.

Naturally the relative errors in the determination of these infinitesimal amounts of radium growing in large volumes of solutions are considerable, and the difficulties are greatly increased by the length of time over which the measurements have extended. The chief sources of error are in the possible change of the constant of the instrument, the change in the standards used to calibrate the instrument, and the actual errors of determination which apply equally to the calibration of the instrument as to the tests on the preparations themselves. Naturally, if the measurements were restarted now, it would be possible, with the greater knowledge and experience now available, to improve the former results. The difficulty is always to be sure that the measurements done, say, five years ago are in every way comparable with those now being done. In future it is proposed to avoid the use of liquid emanation standards, prepared from uranium minerals, altogether.

The quantity of radium in a pure solid barium chloride preparation containing about 6×10^{-10} g. of radium per gram is being determined once for all, and in future fresh weighed quantities of 0.1 to 0.5 gram of this preparation will be used to calibrate the instrument as required. In this way, the measurements over long periods of time may be expected to agree better with one another.

The general character of the results is, however, now fairly clear. As is to be expected, Preparation IV., though the youngest of the four preparations, gives already the most information owing to the very large quantity of uranium—from 8 to 12 times the quantity of any of the other preparations. The growth of radium during the first period of three years from purification was only about one-third the growth in the second period of three years, in agreement with what is to be expected, if the growth in this preparation is entirely due to the uranium and if ionium was initially absent. On this assumption, the period of ionium calculated from the present results agrees fairly well with that calculated for the period on the same assumption three years ago, which indicates that the assumption itself cannot be seriously at fault.

Sir E. Rutherford has shown* that the initial growth of radium from uranium is represented by

$$R = \frac{1}{2} \lambda_2 \lambda_3 R_0 t^2,$$

where R is the number of atoms of radium grown from the number of atoms of uranium in equilibrium with R_0 atoms

* 'Radioactive Substances and their Radiations,' p. 466.

of radium in time t , and λ_2 and λ_3 are the radioactive constants of ionium and radium respectively. The ratio of the mass of radium in equilibrium with 1 gram of uranium is taken throughout this paper to be 3.4×10^{-7} . This factor agrees best with the experiments before referred to, and is somewhat higher than the Rutherford and Boltwood value, recalculated to the International Standard, viz. 3.23×10^{-7} * and in better agreement with the value of Heimann and Marckwald, viz. 3.33×10^{-7} †.

$$\text{Hence } R_0 = 3.4 \times 10^{-7} \times 238/226 \times P,$$

where P is the number of uranium atoms experimented upon. If M is the mass of radium formed from a mass U of uranium,

$$M/U = R/P \times 226/238,$$

and

$$R/P = \frac{1}{2} \lambda_2 \lambda_3 t^2 \times 3.4 \times 10^{-7} \times 238/226.$$

$$\text{Hence } M/U = \frac{1}{2} \lambda_2 \lambda_3 t^2 \times 3.4 \times 10^{-7}.$$

If $1/\lambda_3$ is 2375 years ‡,

$$1/\lambda_2 = (7.16 \times 10^{-11}) U t^2 / M.$$

For Preparation IV., taking the mean of the first two and last two measurements given in the Table (p. 215), when t was 0.26 M was 41, and when t was 5.85 M was 116.5 ($\times 10^{-12}$ g.). Hence

$$\begin{aligned} 1/\lambda_2 &= 7.16 \times 10^{-11} \times 3000 \times (5.85^2 - 0.26^2) / 75.5 \times 10^{-12} \\ &= 97,000 \text{ years.} \end{aligned}$$

In previous calculations the factor 6 instead of 7.16, deduced above, has been used in these calculations. The old factor would make the period 81,000 years if used above. With this may be compared the previously published 70,000 years, deduced three years ago from this experiment as the minimum period of ionium. The curve drawn through the observations on Preparation IV. in fig. 1 is the theoretical curve deduced from the above equation, taking $1/\lambda_2$ as 100,000 years in the above equation. It agrees fairly well with the experimental observations.

Of the other preparations only Preparation III. can yet give any information. In this the initial quantity of radium was excessively minute and the greater relative accuracy of the measurements, in consequence, and the greater age in part compensate for the smallness of the quantity of uranium,

* Sir E. Rutherford, *Phil. Mag.* [6] xxviii. p. 323 (1914).

† Heimann and Marckwald, *Physikal. Zeitsch.* xiv. p. 303 (1913).

‡ Sir E. Rutherford, *loc. cit.* p. 323.

compared with Preparation IV. The curve in fig. 1 for this preparation is the theoretical curve drawn on the same assumption as those for the curve of Preparation IV. As before, taking the mean of the first two and last two observations in the Table (p. 215),

$$\begin{aligned} 1/\lambda_2 &= 7.16 \times 10^{-11} \times 408 \times (8.325^2 - 1.795^2) / 18.5 \times 10^{-12} \\ &= 104,000 \text{ years,} \end{aligned}$$

a value agreeing well with that given by Preparation IV., and with 96,000 years calculated three years ago for this preparation. But in this case the intermediate observations lie consistently above the theoretical curve. The departure amounts, as a rule, to less than 3×10^{-12} g. of radium, and it would be unwise at the present stage to lay too much stress upon it. If it is real, it indicates that the true period of ionium is somewhat longer than that calculated and that in both Preparations III. and IV. some ionium was initially present. From the results with the earlier Preparations I. and II., where certainly some ionium was initially present, nothing can yet be deduced as to the period of ionium. It may be stated in conclusion that the period of average life of ionium is probably about 100,000 years on the assumption that that of radium is 2375 years. This value still partakes of the nature of a minimum period, but it is unlikely that it is very far from the true period.

Summary.

The continuation of the measurements on the growth of radium from purified uranium preparations has shown an unmistakable increase in the rate of growth of radium in the case of the preparation containing 3 kilograms of uranium. The growth of radium appears to be proceeding according to the square of the time, as theory requires if ionium is the only long-lived intermediate member of the series. There is thus now, for the first time, direct experimental evidence that uranium is the ultimate parent of radium. The period of average life of ionium calculated from this experiment is about 100,000 years, assuming 2375 years as the period of radium. An earlier preparation containing 408 grams of uranium gives practically the same value for the period of ionium, calculated on the assumption that ionium was initially absent. The effect of any ionium initially present in the preparations would be to lengthen the period of ionium, but 100,000 years is probably not far from the actual period of average life.