

THE

# AMERICAN JOURNAL OF SCIENCE

[FOURTH SERIES.]

---

ART. VII.—*On the Ultimate Disintegration Products of the Radio-active Elements. Part II. The Disintegration Products of Uranium*; by BERTRAM B. BOLTWOOD.

[Contributions from the Sloane Physical Laboratory of Yale University.]

THE general question of the nature of the ultimate disintegration products of the radio-active elements, as indicated by the occurrence of certain chemical elements in the radio-active minerals, has been discussed in an earlier paper,\* and it was there pointed out that lead, bismuth and barium might perhaps be included among the possible disintegration products. As more recent experiments† have indicated, however, that actinium is probably an intermediate product between uranium and radium, the number of possible ultimate products has been correspondingly reduced. In addition to this careful examinations have been made of specially selected samples of typical primary uraninites from Branchville, Conn., and Flat Rock, N. C., and of thorianite from Ceylon, which have led to the conclusion that neither bismuth nor barium can be considered as disintegration products in the main line of descent from either uranium or thorium, at least on the basis of the present disintegration theory.

The conditions essential for the identification of the final disintegration products of uranium from a study of the composition of the natural minerals which contain this element would appear to be the following: In unaltered primary minerals of the same species, and of different species from the same locality, that is, in minerals formed at the same time and therefore of equal ages, a constant proportion must exist between the amount of each disintegration product and the

\* This Journal, xx, 253, 1905.

† Ibid., xxii, 537, 1906.

amount of the parent substance with which it is associated. And, in unaltered, primary minerals from different localities, the proportion of each disintegration product with respect to the parent substance must be greater in those minerals which are the older and should correspond with the order of the respective geological ages of the localities in which the minerals have been found. It also follows that in secondary minerals, namely, in minerals which have been formed by the subsequent alteration of the original, primary minerals, the relative amounts of the disintegration products must be less than in the primary minerals from the same locality, provided, however, that the disintegration products can not be considered as original chemical constituents of the secondary mineral.

It is the purpose of the present paper to show that the above requirements are practically fulfilled by lead and by helium also, in so far as the gaseous nature of the latter element will permit of its retention in the minerals. The suggestion that lead was one of the final (inactive) disintegration products of uranium was first made by the writer in a paper presented before the New York Section of the American Chemical Society on February 10, 1905, and published later in the *Philosophical Magazine*.\*

The amounts of uranium and lead present in a considerable number of primary uranium minerals have been calculated from the published analyses of these minerals. The number of such analyses to be found in the literature is not large, and, what is still more unfortunate, with the exception of those made by Hillebrand and a few others, cannot be considered as particularly accurate. Many of the analyses were made with special objects in view, such as the identification of a given specimen with a species already known or its recognition as a new variety or species. There is also what is perhaps an unfortunate tendency on the part of many mineralogists to carry out an analysis merely for the purpose of assigning to the mineral some definite chemical formula, which often leads to the overlooking or ignoring of a number of the minor constituents. And in addition to this there are also the actual analytical difficulties to be taken into account, which may be very considerable in the case of such minerals as samarskite, fergusonite, euxenite and other minerals containing notable proportions of niobium, tantalum and titanium. Notwithstanding these objections, however, it is necessary to rely very largely on these published analyses, for the simple reason that the greater number of the uranium minerals are extremely rare and the obtaining of suitable samples of the various species and varieties is either extremely difficult or altogether impossible.

\* April, 1905.

In the table which follows (Table I) are given the results obtained from the calculation of the ratio of the percentage of lead to that of uranium contained in the different minerals as indicated by the analyses.

No.	Mineral	Locality	Per cent U	Per cent Pb	Ratio $\frac{\text{Pb}}{\text{U}}$	Analysis by
1.	Uraninite,	Glastonbury, Conn.,	70	2.9	0.041	Hillebrand, this Journal, xl, 384, 1890.
2.	Uraninite,	Glastonbury, Conn.,	70	3.0	0.043	
3.	Uraninite,	Glastonbury, Conn.,	70	2.8	0.040	Hillebrand, <i>l. c.</i>
4.	Uraninite,	Glastonbury, Conn.,	72	3.0	0.042	Hillebrand, <i>l. c.</i>
5.	Uraninite,	Glastonbury, Conn.,	72	2.9	0.040	Hillebrand, <i>l. c.</i>
6.	Uraninite,	Branchville, Conn.,	74	4.0	0.054	Hillebrand, <i>l. c.</i>
7.	Uraninite,	Branchville, Conn.,	75	4.0	0.053	Hillebrand, <i>l. c.</i>
8.	Uraninite,	Branchville, Conn.,	74	4.0	0.054	Hillebrand, <i>l. c.</i>
9.	Uraninite,	Branchville, Conn.,	66	3.5	0.053	From an analysis by the writer.
10.	Uraninite,	Spruce Pine, N. C.,	77	3.9	0.051	
11.	Uraninite,	Spruce Pine, N. C.,	77	4.2	0.055	Hillebrand, <i>l. c.</i>
12.	Uraninite,	Spruce Pine, N. C.,	67	3.3	0.049	From an analysis by the writer.
13.	Uraninite,	Marietta, S. C.,	71	3.3	0.046	
14.	Uraninite,	Llano Co., Tex.,	55	9.4	0.17	Hillebrand, <i>l. c.</i>
15.	Uraninite,	Llano Co., Tex.,	56	9.5	0.17	Hidden and Mackintosh, this Journal, xxxviii, 481, 1889.
16.	Mackintoshite,	Llano Co., Tex.,	19	3.4	0.18	
17.	Yttrocraosite,	Eurnet Co., Tex.,	2.3	0.44	0.19	Hillebrand, this Journal, xlvi, 98, 1893.
18.	Samaraskite (?)	Douglas Co., Colo.,	3.5	0.67	0.19	Hidden and Warren, this Jour., xxii, 515, 1906.
19.	Samaraskite (?)	Douglas Co., Colo.,	3.7	0.74	0.20	Hillebrand, Proc. Col. Sc. Soc., iii, 38, 1888.
20.	Samaraskite (?)	Douglas Co., Colo.,	5.1	0.99	0.19	Hillebrand, <i>l. c.</i>
21.	Uraninite,	Anneröd, Norway,	66	8.4	0.13	Hillebrand, this Journal, xl, 384, 1890.

80 *B. B. Boltwood—Ultimate Disintegration Products*

No.	Mineral	Locality	Per cent U	Per cent Pb	Ratio $\frac{\text{Pb}}{\text{U}}$	Analysis by
22.	Uraninite,	Anneröd, Nor.,	68	7.8	0.12	Blomstrand, Jour. prakt. Chem., xxix, 191, 1884.
23.	Annerödite,	Anneröd, Nor.,	15	2.2	0.14	Blomstrand, Dana's System of Min., p. 741.
24.	Uraninite,	Elvestad, Nor.,	66	9.3	0.14	Hillebrand, this Journal, xl, 384, 1890.
25.	Uraninite,	Elvestad, Nor.,	57	8.0	0.14	Hillebrand, <i>l. c.</i>
26.	Uraninite,	Skaartorp, Nor.,	65	8.8	0.13	Hillebrand, <i>l. c.</i>
27.	Uraninite,	Huggenäskilen, Nor.,	68	8.8	0.13	Hillebrand, <i>l. c.</i>
28.	Uraninite,	Huggenäskilen, Nor.,	76	9.0	0.12	Lorenzen, Nyt. Mag., xxviii, 249, 1884.
29.	Thorite,	Hitterö, Nor.,	8.2	1.2	0.14	Lindström, G. För. Förh., v, 500, 1881.
30.	Uraninite,	Arendal, Nor.,	56	9.8	0.17	Hillebrand, <i>l. c.</i>
31.	Uraninite,	Arendal, Nor.,	61	10.2	0.17	Hillebrand, <i>l. c.</i>
32.	Uraninite,	Arendal, Nor.,	56	9.4	0.17	Lindström, Zeit. f. Kryst., iii, 201, 1878.
33.	Thorite,	Arendal, Nor.,	9.0	1.5	0.17	Nordenskiöld, G. För. Förh., iii, 228, 1876.
34.	Orangite,	Landbö, Nor.,	7.5	1.2	0.16	Hidden, this Journal, xli, 440, 1891.
35.	Xenotime,	Naresto, Nor.,	2.9	0.62	0.21	Blomstrand, G. För. Förh., ix, 185, 1887.
36.	Hielmite,	Falun, Sweden,	1.9	0.20	0.10	Weibull, <i>ibid</i> , ix, 371, 1887.
37.	Polycrase,	Slättakra, Sweden,	7.4	0.85	0.12	Blomstrand, Dana's Min., p. 745.
38.	Thorianite,	Sabaragamuwa Province, Ceylon,	9.8	2.1	0.21	Dunstan and Blake, Proc. Roy. Soc. Lond., lxxvi (A), 253, 1905.
39.	Thorianite,	Sab. Prov., Cey.,	10.8	2.7	0.25	Dunstan and Blake, <i>l. c.</i>
40.	Thorianite,	Sab. Prov., Cey.,	12.8	2.4	0.19	Dunstan and Blake, <i>l. c.</i>
41.	Thorianite,	Sab. Prov., Cey.,	11.2	2.7	0.24	Analysis by writer.
42.	Thorianite,	Cey.,	11.1	2.3	0.21	Büchner, Nature, lxxv, 169, 1906.
43.	Thorianite,	Galle District, Cey.,	25	2.1	0.086	Dunstan and Jones, Proc. Roy. Soc., Lond., lxxvii (A), 546, 1906.

In the above table the minerals can be divided into seven general groups according to the localities from which they were obtained, namely; Group I from Connecticut (Nos. 1 to 9); Group II from North and South Carolina (Nos. 11 to 13); Group III from Texas (Nos. 14 to 17); Group IV from Colorado (Nos. 18 to 20); Group V from Norway (Nos. 21 to 35); Group VI from Sweden (Nos. 36 and 37); Group VII from Ceylon (Nos. 38 to 43). These groups can be further subdivided into Group I<sub>1</sub> from Glastonbury\* and Group I<sub>2</sub> from Branchville, Group V<sub>1</sub> from localities in the neighborhood of Moss (21 to 29) and Group V<sub>2</sub> from Arendal, and Group VII<sub>1</sub> from the Sabaragamuwa Province and Group VII<sub>2</sub> from the district of Galle.

If the ratio of the lead to the uranium in these groups is now considered, it is evident that in Group I<sub>1</sub> the average value is 0.041 and the maximum divergence is less than five per cent. In Group I<sub>2</sub> the average is 0.0535, which is in close agreement with the four separate values given. In Group II the agreement of the different values is not so good, but is still very striking when the fact is taken into account that the two specimens from North Carolina examined by Hillebrand showed unmistakable evidence of slight secondary alteration as did also the specimen from South Carolina, which moreover is from a different locality and is placed with this group only because there are no others with which to compare it. The material used by the writer was to all appearances free from alteration.

In Group III an opportunity is afforded for the comparison of different species from the same locality, for the yttracrasite described by Hidden and Warren was found only just across the Colorado River from the famous Barringer Hill locality which supplied the other Texas specimens. Here the agreement of the ratios is again very satisfactory, especially in the case of the yttracrasite, which is one of the minerals the care-

\* I have been informed by Mr. E. B. Hurlburt of Glastonbury, Conn., who has made a careful study of the mineral occurrences in his locality, that he considers it to be highly improbable that the specimens examined by Hillebrand and described as from Glastonbury were actually found in that place or even in the neighboring quarries of South Glastonbury. Columbite, monazite, a mineral resembling polycrase and autunite are found at South Glastonbury, but Mr. Hurlburt, who has looked into the matter quite thoroughly, is of the opinion that the specimens of uraninite credited to Glastonbury must have been found in the feldspar quarries of Portland, a town on the east bank of the Connecticut River between South Glastonbury and Middletown. A number of years ago uraninite in some quantity was found at Portland, and as some of the workmen in the Portland quarries had their homes in South Glastonbury, its occurrence in the latter locality may readily have been assumed by the collectors who afterwards obtained the specimens. It is also equally possible that the specimens in many collections labelled as from Middletown are also really from Portland.

ful analysis of which presents so many difficult problems. The writer had the good fortune to meet Professor Warren at the time that this analysis was in progress and the latter kindly consented to take special precautions in the determination of the lead and uranium.

In Group IV the analyses of three samples of a mineral closely resembling samarskite give values for the ratio showing a good agreement, although the different specimens were, according to Hillebrand, quite different in general appearance.

In Group V, the agreement is again very good, while in Group V<sub>2</sub>, including Nos. 34 and 35, the agreement is excellent, for the difference in the case of the xenotime from Narestö, near Arendal, is no more than is to be expected when the relatively small amounts of both uranium and lead are taken into consideration.

In Group VI the ratios given by the two species from different Swedish localities are of little value for the present purposes of comparison, and are significant only as indicating a ratio of the same general order as that found in Group V.

It is unfortunate for the purpose of the present calculation that the analyses of thorianite from Ceylon by Dunstan and Blake and by Dunstan and Jones have been published in so incomplete a form. This interesting mineral, containing a relatively high proportion of both thorium and helium, affords an exceptional material for the study of radio-active changes, and an accurate knowledge of its general composition would be of much assistance in settling some of the doubtful questions. The published analyses are defective however in the following particulars:—In analyses Nos. I and II\* (Nos. 38 and 39 in Table I) the results as given indicate that all of the uranium is present in the form of uranous oxide (UO<sub>2</sub>), while in analysis No. III (*l. c.*) (No. 40 in Table I) a greater proportion of uranic oxide (UO<sub>3</sub>) than of uranous oxide is shown. Such an extreme variation in composition is not only highly improbable, but (in the light of a more recent analysis of thorianite of a similar variety† in which the uranium is given as UO<sub>2</sub>+UO<sub>3</sub> = 13.4 per cent) is probably quite misleading as to the actual composition. Out of the seven analyses given in the paper by Dunstan and Jones, in only one (No. 43 in Table I) are the separate amounts of uranous and uranic oxides shown, while in the other six a number representing the sum of the percentages of the two oxides is inserted, which affords no reliable clue as to the amount of either oxide or the amount of uranium itself contained in the mineral.

The following table containing the values given in the paper by Dunstan and Jones would not appear to be contradictory to

\* *Proc. Roy. Soc. Lond.*, lxxvi (A), 253, 1905. † Dunstan and Jones, *l. c.*

the assumption that the lead and uranium are in a constant ratio to one another in the thorianite from the Galle district.

TABLE II.

Sample No.	UO <sub>2</sub> + UO <sub>3</sub>	PbO*
I	32.7	2.56
II	10.3    18.9	2.29
III	28.2	2.29
IV	28.7	2.50
V	27.0	2.99
VI	28.0	2.90

In the paper by Dunstan and Jones an analysis of a specimen of thorianite from the Balangoda district, showing UO<sub>2</sub> + UO<sub>3</sub> = 13.4 per cent and PbO = 2.54, suggests a close agreement of the ratio of lead to uranium in this mineral with the same ratio in the material from the Sabaragamuwa Province.

The minerals given in the preceding table are all primary minerals, in the general sense in which this term is used. In the following table (Table III), the ratio of lead to uranium has been calculated for some secondary minerals from the same localities.

TABLE III.

No.	Mineral	Locality	Per cent U	Per cent Pb	Ratio Pb / U	Analysis by
1.	Uranophane,	Mitchell Co., N. C.,	55	0.56	0.01	Genth, Am. Ch. J., i, 88, 1879.
2.	Uranophane,	Arendal, Nor.,	40	1.6	0.04	Nordenskiöld, G. Förh., vii, 121, 1884.
3.	Thorogummite,	Llano Co., Tex.,	19	2.0	0.10	Hidden and Mackintosh, this Journal, xxxviii, 480, 1889.

These analyses all agree in giving a lower ratio for the secondary minerals than for the primary minerals from the same localities. The most common alteration product of uraninite known as gummite can be left out of present consideration since lead is apparently one of its natural, chemical constituents.

The actual value of the ratio varies considerably for the primary minerals from different localities, the maximum value being about six times the minimum. It is beyond the writer's province to discuss the data bearing on the geological ages of the different deposits, but he is indebted to Professor Joseph Barrell of Yale University for the statement that, so far as the knowledge of the latter extends, the relative values of the

ratios are not contradictory to the order of the ages attributed by geologists to the formations in which the different minerals occur.

From the data which have been presented in the preceding tables it is apparent that the requirements for a disintegration product of uranium are fulfilled by lead within the limits of probable experimental error. On the basis of this evidence the assumption would appear to be justified that lead is the final product of uranium.

#### *Helium.*

Few experimental determinations of the relative quantities of helium in minerals of known composition are to be found in the literature. A careful search has brought to light only the following: Twelve determinations by Hillebrand\* of the "nitrogen" present in an equal number of samples of uraninites of known composition; the determination by Ramsay and Travers† of the per cent of helium in a sample of fergusonite, the analysis but not the locality of which is given; the determinations by Strutt‡ of the amounts of helium in a number of minerals which had been analyzed for uranium only; a determination by Dunstan and Blake§ of the helium in an analyzed sample of thorianite from the Sabaragamuwa province, Ceylon; and the determination of helium in another specimen of the same mineral by Büchner.||

Considering the great exactness of all the analytical work carried out by Hillebrand, and the general method which he followed in his determinations of "nitrogen," it is highly probable that by dividing the values which he gives in his paper by seven ( $N_2 : He = 28 : 4$ ) a very reliable number for the percentage of helium is obtained.

It has been shown conclusively by a number of different experimenters that the disintegration of radium is accompanied by the production of helium, and it is further stated by Debierne¶ that the disintegration of actinium furnishes helium also. If the assumption is made on the basis of analogy that the entire change from uranium to lead is accompanied by the production of helium, then the quantities of matter involved in this change can be represented by the equation

$$\text{Uranium (238.5)} = \text{lead (206.9)} + \text{helium (31.6)},$$

in other words, that for every 207 parts of lead there will be formed 32 parts of helium.

From a knowledge of the amount of lead present in the minerals it is, therefore, possible to calculate the amount of helium

\* This Journal, xl, 384, 1890; *ibid.*, xlii, 390, 1891.

† Proc. Roy. Soc. Lond., lii, 316, 1898. ‡ *Ibid.*, Lond., lxxvi (A), 88, 1905.

§ *l. c.* || Nature, lxxv, 165, 1906. ¶ C. R., cxli, 383, 1905.

which would be formed according to this hypothesis, and to compare this amount with the amount actually present in the minerals.

Calculations and comparisons of this sort have been made for a number of minerals and the results are given in the following table (Table IV).

TABLE IV.

No.	Mineral	Per cent Lead	Per cent Helium		R
			present	calculated	
1.	Uraninite, Glastonbury, Conn.,	2.9	0.34	0.43	79
2.	Uraninite, Branchville, Conn.,	4.0	0.39	0.60	65
3.	Uraninite, Elvestad, Nor.,	9.3	0.18	1.40	13
4.	Uraninite, North Carolina,	3.9	0.05	0.58	9
5.	Uraninite, Skaartorp, Nor.,	8.8	0.15	1.32	11
6.	Uraninite, Huggenäskilen, Nor.,	8.8	0.15	1.32	11
7.	Uraninite, Anneröd, Nor.,	8.4	0.17	1.26	13
8.	Uraninite, Elvestad, Nor.,	8.0	0.15	1.21	12
9.	Uraninite, Llano Co., Tex.,	9.4	0.08	1.40	6
10.	Uraninite, Colorado,	0.6	0.02	0.10	20
11.	Uraninite, Arendal, Nor.,	10.2	0.16	1.53	10
12.	Thorianite, Ceylon,	(2.6)	0.16	0.40	40
13.	Aeschynite, Hitteroe, Nor.,	(1.2)	0.02	0.18	11
14.	Samarskite, North Carolina,	(0.42)	0.03	0.06	50
15.	Gadolinite (?), Ytterby, Sweden,	(0.25)	0.04	0.04	100
16.	Cyrtolite, Texas,	(0.53)	0.02	0.08	25
17.	Euxenite, Arendal, Nor.	(0.41)	0.013	0.06	20
18.	Uraninite, Canada,	10.5	0.12	1.6	8
19.	Thorianite, Ceylon,	2.40	0.19*	0.36	53
20.	Thorianite, Ceylon,	2.25	0.15	0.34	44

Nos. 1 to 11 and No. 18 are from analyses by Hillebrand, Nos. 12 to 17 from determinations by Strutt (the per cent of lead being calculated from the ratios in Table I), No. 19 from the analysis of Dunstan and Blake, and No. 20 from that of Büchner. In the last column under the heading *R* is given the ratio of the amount of helium actually present to the amount formed according to the hypothesis multiplied by 100, or in other words, the percentage of the total helium formed which has been retained by the mineral.

It will be noted that the values obtained in this manner for *R* are very reasonable numbers and are not unlike what might be expected from general considerations. This relation is the more evident when the density of the minerals is also exam-

\* In the analysis of Dunstan and Blake an error is made in calculating the per cent of helium in this mineral, which is given as 0.39 per cent of helium. It is stated that the mineral contained 10.5<sup>cc</sup> of helium per gram which is equal to 0.19 per cent of helium.

ined. The densities of only the first ten minerals are known, and these densities with the corresponding values for  $R$  are given in the table which follows.

TABLE V.

No.	Sp. gr.	R	No.	Sp. gr.	R
1.	9.62	79	6.	8.93	11
2.	9.35	65	7.	8.89	13
3.	9.14	13	8.	8.32	12
4.	9.08	9	9.	8.29	6
5.	8.96	11	10.	8.07	20*

None of the minerals listed in Table IV contains *more* helium than is to be expected from the assumption that helium is produced by the disintegration of uranium only, and in general with greater density of the mineral a greater proportion of the total helium formed has been retained within it.

#### *Age of Minerals.*

If the quantity of the final product occurring with a known amount of its radio-active parent and the rate of disintegration of the parent substance are known, it becomes possible to calculate the length of time which would be required for the production of the former. Thus, knowing the rate of disintegration of uranium, it would be possible to calculate the time required for the production of the proportions of lead found in the different uranium minerals, or in other words the ages of the minerals.

The rate of disintegration of uranium has not as yet been determined by direct experiment, but the rate of disintegration of radium, its radio-active successor, has been calculated by Rutherford† from various data. Rutherford's calculations give 2600 years as the time required for half of a given quantity of radium to be transformed into final products. The fraction of radium undergoing transformation per year is accordingly  $2.7 \times 10^{-4}$ , and preliminary experiments by the writer on the rate of production of radium by actinium‡ have given a value which is in good agreement with this number. The quantity of radium associated with one gram of uranium in a radio-active mineral has also been determined§ and was found to be  $3.8 \times 10^{-7}$  gram. On the basis of the disintegration theory, when radium and uranium are in radio-active equilibrium, an equal number of molecules of each disin-

\* It should be explained that No. 10 is really a secondary uraninite and is, therefore, not directly comparable with the others.

† Phil. Mag. (6), xii, 367, 1906.

‡ This Journal, xxii, 537, 1906.

§ Rutherford and Boltwood, this Journal, xxii, 1, 1906.

tegrate per second, and, for our present purposes, we can neglect the difference in atomic weight and simply assume that in any time the weights of radium and uranium which undergo transformation are the same. In one gram of uranium the weight of uranium which would be transformed in one year would therefore be  $2.7 \cdot 10^{-4} \times 3.8 \cdot 10^{-7} = 10^{-10}$  gram, and the fraction of uranium transformed per year would be  $10^{-10}$ .

In the table which follows (Table VI) the ages of the minerals included under Table I have been roughly calculated in accordance with the method outlined above. The ages of the minerals in years are obtained by multiplying the average value of the ratio  $10^{10}$ . The general plan of calculating the ages of the minerals in this manner was first suggested to the writer by Prof. Rutherford.

TABLE VI.

Locality.	Age of minerals in million years.
Glastonbury (Portland), Conn. ....	410
Branchville, Conn. ....	535
Spruce Pine, N. C. ....	510
Marietta, S. C. ....	460
Llano and Burnet Co., Texas. ....	1800
Douglas Co., Colorado ....	1900
Moss District, Norway ....	1300
Annerod, Norway ....	1700
Sabaragamuwa Prov., Ceylon ....	2200
Galle District, Ceylon ....	860

The actual values obtained for these ages are, of course, dependent on the value taken for the rate of disintegration of radium. When the latter has been determined with certainty, the ages as calculated in this manner will receive a greater significance, and may perhaps be of considerable value for determining the actual ages of certain geological formations.

#### *Disintegration Products of Thorium.*

The available data on the composition of the radio-active minerals serve to throw some light on the nature of the disintegration products of thorium as well as uranium. The relative proportions of uranium and thorium may show large variations in minerals from the same locality without exercising a noticeable effect on the value of the lead-uranium ratio for that locality. It can therefore be concluded with certainty that lead is not a disintegration product of thorium. This fact is particularly emphasized by the composition of the thorite found with the thorianite in the Sabaragamuwa province of Ceylon and in all probability of contemporaneous

formation. The constituents of this mineral\* are in part as follows:  $\text{ThO}_2$ , 66.26 per cent;  $\text{CeO}_2$ , 7.18 per cent;  $\text{ZrO}_2$ , 2.23 per cent;  $\text{UO}_2$ , 0.46 per cent. No lead at all is indicated as present, and the amount to be expected from the uranium is only 0.08 per cent, which was probably overlooked in making the analysis. A similar result was obtained in an examination by the writer of a specimen of thorite from Norway, which contained only 0.40 per cent of uranium, 52.0 per cent of  $\text{ThO}_2$  and less than 0.10 per cent of lead. No mention is made of the presence of helium in the former of these thorites and in the specimen examined by the writer no indications of the presence of helium in measurable quantities were obtained. Although it has been stated by Ransay† that the relatively large amount of helium contained in the thorianite from the Sabaragamuwa province is conclusive evidence of the production of helium by thorium, it seems quite probable that the evidence furnished by this mineral is quite the contrary, since it appears to contain only half of the amount of helium which would be produced by the disintegration of the uranium alone.

#### *Summary.*

Evidence has been presented to show that in unaltered, primary minerals from the same locality the amount of lead is proportional to the amount of uranium in the mineral, and in unaltered primary minerals from different localities the amount of lead relative to uranium is greatest in minerals from the locality which, on the basis of geological data, is the oldest. This is considered as proof that lead is the final disintegration product of uranium.

It has also been shown that, on the basis of the experimental data at present available, the amounts of helium found in radio-active minerals are of about the order, and are not in excess of the quantities, to be expected from the assumption that helium is produced by the disintegration of uranium and its products only.

The improbability that either lead or helium are disintegration products of thorium has been pointed out.

December 27, 1906.

\* Dunstan and Blake, *l. c.*

† Jour. Chem. Phys., iii, 617, 1905.