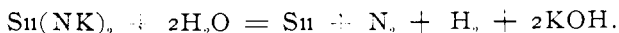
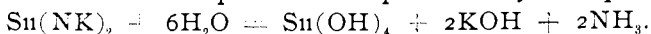


tion that a portion of the salt was decomposed in accordance with the equation,



This partial decomposition is to be ascribed to the violence of the action of liquid water and the high temperature thereby produced. The hydrolytic action of the water vapor is to be represented by the equation,



#### Summary of Results.

It is shown by the work described above that a potassium ammonostannate of the formula,  $\text{Sn}(\text{NK})_2 \cdot 4\text{NH}_3$ ,  $(\text{NH}_2)_2:\text{Sn}:(\text{NKH})_2 \cdot 2\text{NH}_3$ ,  $(\text{KHN})_2:\text{Sn}:(\text{NHNH}_4)_2$ , or  $\text{Sn}(\text{NH}_2)_4 \cdot 2\text{KNH}_2$ , an ammonia analogue of potassium stannate, is formed when a solution in liquid ammonia of potassium amide in excess is allowed to act upon stannic iodide, likewise in solution in liquid ammonia, and that this compound, when heated to a temperature not in excess of  $145^\circ$ , loses three molecules of ammonia of crystallization to form the compound,  $\text{Sn}(\text{NK})_2 \cdot \text{NH}_3$  or  $\text{HN}:\text{Sn}:(\text{NKH})_2$ . From this compound even by heating for several hours *in vacuo* to  $316^\circ$  it was not possible to obtain the ammonia-free salt.

The writer is under obligations to Dr. E. C. Franklin of Stanford University for his many suggestions and great assistance in carrying on this investigation.

STANFORD UNIVERSITY,  
California, Summer 1907.

[CONTRIBUTION FROM KENT CHEMICAL LABORATORY, UNIVERSITY OF CHICAGO.]

### THE SPECIFIC RADIOACTIVITY OF URANIUM.<sup>1</sup>

BY HERBERT N. MCCOY AND W. H. ROSS.

Received October 14, 1907.

It may be considered as fully established<sup>2</sup> that the specific activity of Uranium in any pure uranium compound or in any uranium mineral, free from thorium, is in either case strictly constant. The value of these constants has been given as 790 for pure compounds and 3280 for minerals. These values were obtained from activity measurements made with a distance of 3.5 to 4.5 cm. between the active film and the charged electrode of the gold leaf system. It is now known<sup>3</sup> that the ranges of RaA and RaC both exceed 4.5 cm.; for which reason the value given for the specific activity of uranium in a mineral is too low. New measurements have now been made by means of a new improved gold-leaf electroscope which has an ionization chamber 19.5 cm. square and allows a distance of 8.5 cm. between the active films and the superimposed

<sup>1</sup> Read, in abstract, before the Amer. Phys. Soc., Chicago, Dec. 1, 1906; *Phys. Rev.*, Jan., 1907.

<sup>2</sup> McCoy, *Ber.*, 37, 2641 (1904); *This Journal*, 27, 391 (1905); *Phil. Mag.*, 11, 176 (1906); Goettsch, *This Journal*, 28, 1541 (1906).

<sup>3</sup> Bragg and Kleeman, *Phil. Mag.*, 10, 318 (1905).

electrode. The scale of the micrometer microscope, used to read the position of the gold-leaf, corresponded to potentials between 400 and 480 volts. With this new electroscope the activity of a pure uranium compound was the same at 4.5 cm. as at 8.5 cm. from the electrode. But a thin film of a uranium mineral was about 4 per cent. more active at 8.5 cm. than at 4.5 cm.

The recent work on the range of the  $\alpha$  rays has also served to indicate another possible error in the method previously employed for the measurement of activity. According to this method films of the active material were deposited in flat, circular, metal dishes about 7 cm. in diameter, with rims 0.8 cm. high.<sup>1</sup> Inasmuch as the solid angle subtended by the rim, with any point of the film as center, was the same for all films, the rims being of equal height, it was thought, at first, that the same fraction of the radiation would be cut off by the rim in all cases. However, films sending out  $\alpha$ -rays with greatest effective range should suffer greatest proportionate loss of activity on account of the rims. Experiment confirmed this supposition, although the difference in percentage loss was not great. Thus, while the actual diminution of the activity due to the rims was found to be 8.2 per cent for uranium oxide, the corresponding decrease in the case of uranium minerals amounted to 8.8 per cent.; a difference of 0.6 per cent. For the new measurements all films were deposited on perfectly flat surfaces. This was readily done by placing false bottoms, 7.00 cm. in diameter, made of sheet copper, in the tin vessels previously used. After the deposition of the film by the sedimentation process<sup>2</sup> upon the false bottom, the latter was removed from the tin vessel. Perfectly satisfactory films, weighing as little as one milligram per sq. cm., were easily made in this way.

The standard of activity was a thick film of the oxide  $U_3O_8$ , weighing about 0.025 g. per sq. cm. It was deposited on a flat copper plate 7.00 cm. in diameter. The activity due to each sq. cm. of such a film is taken as the unit of activity. The total activity of 1 g. of any substance, in terms of such a unit, may be called its specific activity.

The specific activity of pure uranium oxide,  $U_3O_8$ , was redetermined from measurements of a series of new films on flat plates, the new electroscope being used with the films at 8.5 cm. from the electrode. The results, calculated by the method of graphical extrapolation previously used<sup>3</sup> gave for the specific activity of the oxide, 676. Since the oxide contains 84.85 per cent. of uranium, the specific activity of uranium is 796. This value differs but little from the value, 790, found earlier.

For the new experiments upon minerals, new samples were made from

<sup>1</sup> Tin-plate jelly-glass covers were used.

<sup>2</sup> McCoy, *This Journal*, 27, 395 (1905).

<sup>3</sup> *Phil. Mag.*, 11, 176 (1906).

the three lots of pitchblende previously studied. In the analysis the uranium was separated essentially by Boltwood's modification<sup>1</sup> of Brearley's method. The uranium phosphate so obtained was ignited and weighed. The pyrophosphate was then dissolved in dilute sulphuric acid, reduced with zinc and titrated with permanganate. The volumetric results agreed with the gravimetric very closely. Duplicate or triplicate analyses were made of each sample. All of the samples were analyzed for thorium by Neish's method<sup>2</sup>, 5 gram portions of each mineral being taken, but no trace of thorium was found in any case.

TABLE I.

	Locality.	Per cent. of Uranium.	Specific Activity, k <sub>r</sub>
1.	Pitchblende	Unknown	44.7
2.	"	Central City, Colorado	54.0
3.	"	Gilpin Co., "	58.1
			1582
			1908
			2018

In order to obtain exact results, the specific activities given above must be corrected for the activity of the emanation spontaneously lost; for, as Boltwood has shown<sup>3</sup> all minerals give off a portion of their emanation when powdered. The amount of emanation spontaneously lost was determined essentially in the manner described by Boltwood, the emanation being allowed to accumulate in maximum amount. The emanation was removed from the tube containing the known weight of powdered mineral, by drawing through the former about 250 cc. of air. This was collected in a flask and allowed to stand fifteen minutes before being conducted into the exhausted electroscope. The activity was measured after the emanation had remained in the electroscope two hours, at the end of which time the activity had become practically constant.

In the determination of the emanation retained in a powdered mineral, we have found it possible to substitute for the complicated apparatus and procedure employed by Boltwood, a very simple method. An accurately weighed portion of the mineral, about 0.03 g., was placed in a 150 cc. flask fitted with a rubber stopper and bent glass tube. The latter was connected by a rubber tube to another bent glass tube which just passed through the stopper of a liter flask filled with dilute sodium hydroxide solution. A second tube passed through the stopper of the liter flask and formed a siphon leading from the bottom of the flask emptying into a large beaker containing a little water. The process consisted in pouring onto the mineral in the smaller flask a mixture of 15 cc. of dilute sul-

<sup>1</sup> Phil. Mag., 9, 603. We found it necessary to alter the proportions of the reagents suggested by Boltwood. The details of our procedure are given elsewhere. W. H. Ross, Thesis, Univ. Chicago, 1907.

<sup>2</sup> This Journal, 26, 780 (1904). We have thoroughly tested Neish's method in connection with studies on thorium minerals and found it remarkably accurate.

<sup>3</sup> Loc. cit.

phuric and 5 cc. of nitric acids, inserting the stopper and boiling gently for 10 to 15 minutes. The air originally in the small flask and the gases formed by the reaction were carried by the steam into the liter flask. Once started, the process goes on smoothly and requires no troublesome manipulation. The emanation was allowed to stand in the liter flask 15 minutes before being transferred to the electroscope. The sum of the activities of the evolved and retained emanation is the activity of the total emanation.

In the following table the activities are expressed in arbitrary units, for 1 g. of the mineral. The value given in each case is the mean of two to four concordant determinations.

TABLE 2.

	Activity of retained Emanation	Activity of evolved Emanation	Activity of total Emanation	Per cent. of total Emanation Evolved	Per cent. of Uranium	Activity of per gram Uranium
1	62.8	2.46	65.4	3.76	44.7	146.0
2	...	6.49	78.6	8.25	54.0	145.6
3	80.0	3.69	83.7	4.41	58.1	144.0

The last column shows a close proportionality between the total amount of emanation and the percentage of uranium in the mineral, as previously shown by Boltwood.

The retained emanation of Sample No. 2 could not be determined in the manner described, owing to the fact that the mineral was not completely disintegrated by treatment with boiling dilute sulphuric and nitric acids.

The sample was easily brought into solution after fusion with potassium acid sulphate. The solution obtained from a known weight of mineral was placed in a small flask, boiled to expel all emanation present, and then sealed up. At the end of a known period of time (about 4 days), the emanation which had formed was boiled off, collected, and its activity determined. The activity of the total emanation was thus calculated by means of the equation,  $I_0 = Ie^{\lambda t}$ , where  $\lambda$  was taken as 0.173, corresponding to a period of 3.99 days.<sup>1</sup>

It is probable that the emanating power of a compact mineral like pitchblende is negligibly small until the mineral is powdered. If so, the loss of activity must date from the time when the mineral is pulverized. During a few weeks, or even months, after pulverization, loss of emanation will therefore cause an appreciable deficiency in the amounts of the products of rapid decay, A, B and C, only; while the amount of D, period 40 years, will not be noticeably altered. The amounts of E and F which are products of D will also be practically unchanged by loss of emanation. In order that one may know by what fraction the specific activity of a mineral is decreased by the spontaneous loss of emanation after pulverization,

<sup>1</sup> Curie, Compt. rend., 135, 857 (1902).

it is necessary to know what fraction of the total activity is due to  $E_m + A + B + C$ . The experimental determination was made as follows: A portion of sample No. 1, finely ground was treated with nitric acid, evaporated to dryness and the process repeated three times at intervals of two hours, in order to free the material from  $E_m$  and allow A, B and C to decay. After the last evaporation, the residue was heated just strongly enough to decompose the nitrates present. The resulting residue, consisting essentially of oxides, was made into films as quickly as possible, and its activity determined in the usual way. Two closely agreeing experiments showed that 52 per cent. of the original activity of the mineral had been removed by the treatment. Therefore the activity due to  $E_m + A + B + C$  is approximately half the total activity of the mineral.

A similar conclusion is also reached from the following theoretical considerations. Boltwood has shown<sup>1</sup> by direct experiment that the activity of  $E_m + A + B + C$  is 4.64 times as great as that of the equilibrium amount of radium free from its products. We find, as shown in the following paragraph, that radium free from its products is 0.53 as active as the equilibrium amount of uranium in a mineral, and, also that the activity of a mineral is 4.54 times that due to the uranium alone. There-

fore the activity of  $E_m + A + B + C$  in a mineral is  $\frac{4.64 \times 0.53}{4.54}$

0.54 of the total activity. Direct experiment gave 0.52.

#### The Specific Activity of Uranium in Minerals.

The proper corrections may now be applied to the observed specific activities of the minerals as given in Table 2. The results are given in Table 3.

TABLE 3.

Sample	$k_1$ Uncorrected	Per Cent. of $E_m$ Evolved	Per Cent. of Activity Lost	$k_1$ Corrected	Amount of Uranium Per gram (P)	$\frac{k_1}{P}$
No. 1	1582	3.76	1.9	1613	0.447	3610
No. 2	1908	8.25	4.1	1990	0.540	3685
No. 3	2018	4.41	2.2	2064	0.581	3552
					Mean	3616

The quantity, 3616, is the specific activity of uranium in minerals. The corresponding constant for pure uranium compounds is 796. The ratio of the two quantities is 4.54. For equal uranium content, minerals free from thorium are 4.54 times as active as pure compounds.

Danne has shown<sup>2</sup> that radium occurs in certain uranium free minerals, from Issy-l'Éveque. A specimen of this material was given to us by Prof. Millikan, who had received the sample from Prof. Langevin. We wish to thank both gentlemen for the specimen. We found that a lump

<sup>1</sup> Am. J. Sci., 21, 413 (1906).

<sup>2</sup> Compt. rend., 140, 241 (1905).

of the mineral, which contained no uranium, was more active than an equally large lump of pure pitchblende. But when the lump was powdered and spread over a 20-fold greater surface its activity, instead of being increased about 20-fold, as would have been the case with pitchblende, was decreased 70 per cent! *The activity was almost wholly superficial.* When a lump of the mineral was boiled with dilute hydrochloric acid a few minutes, only traces of the substance went into the solution; but 97 per cent. of the activity was removed. The minute residue left upon evaporation of the acid solution, was highly active. Danne has suggested that the radium has been deposited comparatively recently by percolating subterranean waters; our observations tend strongly to confirm this view. The occurrence of radium in uranium-free minerals does not invalidate the law of fixed proportions of uranium and radium in minerals; since the law applies only to minerals sufficiently old to have reached equilibrium conditions. It only shows that radium may be separated from uranium by natural, as well as by artificial, processes.

We have shown<sup>1</sup> that, when the activity due to uranium is deducted, the remaining activity of any thorium mineral is proportional to its percentage of thorium.<sup>2</sup> It was also shown that the specific activity of thorium containing the equilibrium amounts of its products is the same in pure compounds as it is in minerals; in this respect thorium differs markedly from uranium. Inasmuch as the activity of a thorium mineral is completely accounted for, thorium cannot be the parent of radium. On the other hand it was important to discover to what extent the activity of uranium and radium and their known products would account for the activity of uranium minerals, free from thorium. A summary of our results on this topic has already been published.<sup>3</sup> The experimental process consisted in the determination, firstly, of the activity due to radium, free from its products, in 1 cc. of a solution which contained no other active element and, secondly of the amount of radium emanation contained in 1 cc. of the solution in terms of the amount associated with 1 g. of pure uranium in a mineral. It was thus found that the activity of the radium (free from its products) contained in a mineral is 0.53 of that of the equilibrium amount of uranium.

The radium solution was prepared from the insoluble residue left after the treatment of a good sample of pitchblende with nitric and hydrofluoric acids. One gram of the residue was boiled with 100 cc. of 15 per cent. sodium carbonate solution for 30 minutes. The residue was filtered out and washed and then dissolved in hydrochloric acid. About 0.3 g.

<sup>1</sup> Am. J. Sci., 26, 433 (1906).

<sup>2</sup> Similar results were also announced by Boltwood and by Dadourian in the same number of the Am. J. Sci. as that which contained our paper.

<sup>3</sup> Abstract of a paper read before the Am. Phys. Soc.; Phys. Rev., Jan., 1907.

of bismuth chloride was added. The copper, lead, bismuth, etc., were then precipitated with hydrogen sulphide. The sulphide precipitate was several times as active as uranium and probably contained all of the RaF. The filtrate was boiled to expel hydrogen sulphide completely and then oxidized with bromine. The iron was precipitated with ammonia. The filtrate was treated with a little sulphuric acid and the small precipitate of barium (and radium) sulphate was filtered out. The precipitate was then converted into carbonate and the latter into chloride. The solution of the chloride was evaporated to dryness in a platinum dish, taken up in 200 cc. of water and filtered to remove any possible trace of insoluble matter.

Portions of 5 cc. of the radium solution were evaporated on flat platinum plates 6 cm. square. At intervals of about one hour, the residue was dissolved on the plate in a little dilute hydrochloric acid and again evaporated, in order to remove the emanation and the excited activity. During the fourth (the last) evaporation the solution was so manipulated, by means of a bit of bent platinum wire, that the resulting residue of less than 1 mg. weight, was spread evenly over the whole plate. There is no appreciable absorption of the  $\alpha$ -ray activity in so thin a film. The activity of the film (with the bit of wire) was determined by comparison with a thick film of uranium oxide of known activity. As the mean of a number of such determinations, the total activity of the radium in 5 cc. of the solution was found equal to the total activity of 0.1237 g. of uranium.

A portion of the radium solution was diluted 20-fold and portions equal to 0.5 cc. of the original solution were diluted with water and placed in 150 cc. flasks. The emanation present was completely expelled by boiling and the flasks were then sealed. After known intervals of 4 to 5 days the emanation was boiled off from each solution, its activity determined and the equilibrium amount calculated in the usual way. The equilibrium amount of emanation of 5 cc. of the original radium solution equaled that for 0.2312 g. of uranium in a mineral. The ratio,  $\frac{0.2312}{0.1237} = 1.87$ , represents the relative activity of equilibrium amounts of uranium and radium in a mineral. Conversely radium is 0.53 as active as the equilibrium amount of uranium.

Boltwood has shown<sup>1</sup> that radium plus the maximum amounts of Em, A, B, and C is 5.64 times as active as radium alone. His experiments also indicate, as he pointed out, that the activity of the equilibrium amount of any member of a radioactive series is proportional to its range. The range of the  $\alpha$ -rays of radium F is 3.85 cm.<sup>2</sup> while that of radium is

<sup>1</sup> Am. J. Sci., 21, 409.

<sup>2</sup> Levin, Am. J. Sci., 22, 8 (1906).

3.50 ccm.<sup>1</sup> Therefore F is 1.10 times as active as the equilibrium amount of radium alone. Since D is inactive and E gives only  $\beta$ -rays, uranium together with radium and its products are  $1.87 + 5.64 + 1.10 = 8.61$  times as active as the radium alone; or  $8.61/1.87 = 4.60$  times as active as the equilibrium amount of uranium. Our direct experiments gave 4.54 as the ratio of the activity associated with equal amounts of uranium in minerals and in pure compounds. The close agreement of the two values would seem to indicate that the activity of a uranium mineral (free from thorium) is due solely to uranium and radium and their recognized products.

That radium is a transformation product of uranium is now apparently fully established. The abnormally slow growth of radium in pure uranium<sup>2</sup> is no doubt due to an inactive product of long period compared with radium, between U and Ra. For a time it seemed probable that actinium was this intermediate product; since actinium separated from uranium minerals actually produced radium emanation at approximately the calculated rate<sup>3</sup>.

But Rutherford found later<sup>4</sup> that the rate of growth of radium in preparations obtained from the fractionation of actinium solutions, is not proportional to the quantity of actinium present; and concluded that the parent of radium is only accidentally associated with actinium. There is also another weighty reason against the view that actinium is the product in question. The sum of the ranges of actinium and its products is 22.65 cm.<sup>5</sup> while that of radium is 3.50 cm. Therefore actinium and its recognized products should be  $22.65/3.50 = 6.5$  times as active as radium, or 3.5 times as active as the equilibrium amount of uranium, considering actinium a member of the uranium series. But the observed activity of uranium minerals is insufficient to account for more than a small fraction of this estimated actinium activity. Boltwood has considered this matter<sup>6</sup> and has suggested that each of the other  $\alpha$ -ray changes in the uranium-radium series takes place with the expulsion of four  $\alpha$ -particles. Such a view is interesting, but not in harmony with a number of facts, as Boltwood himself admits.

The fact that the activity of uranium ores seems to be fully accounted for by the activity of uranium and radium and their known products,

<sup>1</sup> Bragg and Kleeman, Loc. cit.

<sup>2</sup> Boltwood, Am. J. Sci., 20, 239 (1905); Soddy, Nature, 76, 150 (1907).

<sup>3</sup> Boltwood, Am. J. Sci., 22, 537 (1906); Rutherford, Nature, 75, 270 (1907).

<sup>4</sup> Nature, 76, 126 (1907).

<sup>5</sup> Hahn, Phil. Mag., 12, 253 (1906).

<sup>6</sup> Nature, 75, 223 (1907). In this "Letter to the Editor" which contains no account of experimental details, Dr. Boltwood states that radium is 0.52 as active as the equilibrium amount of uranium; we found 0.53. He also states that minerals are 5.3 times as active as pure uranium compounds of the same uranium content. This is much greater than the value, 4.54, which we find for the same ratio.



indicates that but a very small fraction of the activity of a mineral can be due to actinium. If we adhere to the views that but one  $\alpha$ -particle is expelled from an atom during a single change, and that activity in any series is even approximately proportional to range; then we must conclude that the number of atoms of actinium changing per second, in a given mass of a mineral, is far less than the number of atoms of uranium changing.

Of course, actinium may not be formed from uranium, but its presence in uranium minerals seems to indicate a genetic relationship. The following hypothesis offers a plausible explanation which is in harmony with the known facts. It is conceivable that atoms of UX may decompose in two ways, each with its own rate of change (such cases are frequent in ordinary chemical decompositions). The more rapid disintegration may yield the radium series; while the other slower disintegration gives the actinium series. If the period of the second sort of change of UX is, say, 0.01 of that of the first; then, for every 101 atoms of uranium changed, one atom of actinium would be formed and disintegrated. Therefore the total activity of actinium and its products would be but a fraction of one per cent. of the total activity of a mineral.

There is perhaps a little evidence that UX does change in two ways. Moore and Schlundt<sup>1</sup> observed that UX apparently has faint  $\alpha$ -ray activity which decays with the same period as the  $\beta$ -activity. Levin<sup>2</sup> has confirmed Moore and Schlundt's observations, as has also Mr. G. C. Ashman, working in this laboratory. But Levin has shown that a large part of the so-called  $\alpha$ -ray activity is probably due to secondary  $\beta$ -rays. Still, according to Levin, 8 per cent. of the activity of UX may be due to  $\alpha$ -rays. Now it is possible that the change involving the expulsion of a  $\beta$ -particle produces, eventually, radium and that giving an  $\alpha$ -particle produces actinium. The faint  $\alpha$ -ray activity of UX may, according to this view, be due to an abnormally small number of  $\alpha$ -particles produced, rather than to a very short range of particles expelled in normal numbers.

As has been stated, uranium is 1.87 times as active as the equilibrium amount of radium; since the range of the latter is 3.50 cm., that of the former should be  $1.87 \times 3.50 = 6.65$  cm., if we assume the range proportional to the activity. This estimated range is about twice as great as one would expect, considering Rutherford's experiments on the thickness of air through which the  $\alpha$ -rays of uranium, radium or thorium pass before being reduced in intensity to half value<sup>3</sup>. We have made some measurements of the range of uranium essentially according to the method of Bragg and Kleeman<sup>4</sup> using a very uniform film of  $U_3O_8$ , made by the

<sup>1</sup> Phil. Mag., 12, 393 (1906).

<sup>2</sup> Physik., 8, 129 (1907).

<sup>3</sup> Rutherford, Radioactivity, 168 (1903).

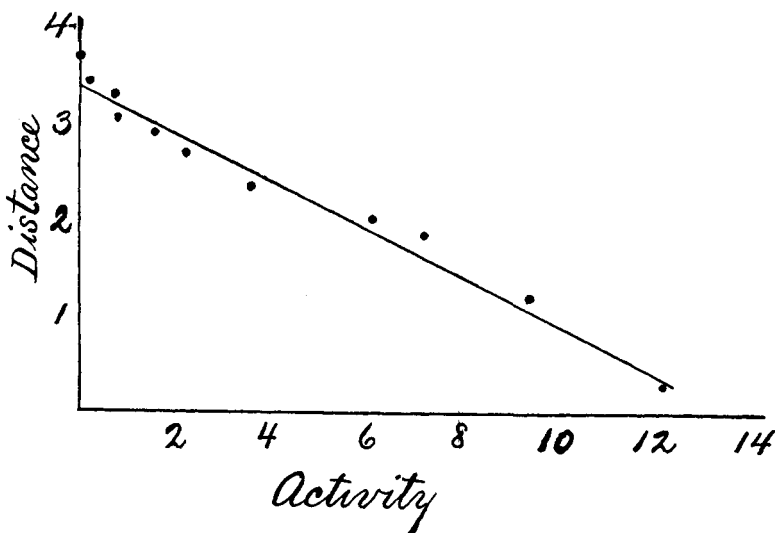
<sup>4</sup> Loc. cit.

usual process of sedimentation. The film was 7 cm. in diameter and weighed 43 mg. It was enclosed in a copper cylinder 8 cm. in diameter and 5 cm. high with a wire gauze top and solid base. The film could be placed at any desired distance below the gauze, which was fixed at a distance of 0.5 cm. below the electrode attached to the gold leaf system. The following table gives the observed activities, in arbitrary units for various distances in cm., between the film and the gauze.

TABLE 4.

Distance	0.30	1.20	1.85	2.00	2.35	2.70	2.90	3.05	3.30	3.45	3.70
Activity	12.2	9.4	7.2	6.1	3.8	2.2	1.6	0.8	0.7	0.2	0.0

The results are reproduced graphically in the accompanying figure.



The low specific activity of uranium made it impossible to employ extremely thin films such as Bragg and Kleeman used in determining the range of radium. On account of the appreciable thickness of the uranium film used, the  $\alpha$ -ray radiation was not of uniform range, and so did not give results like those obtained with infinitely thin films. Nevertheless the experiment seems to show that the maximum range of uranium is about 3.4 cm. Bragg's experiments<sup>1</sup> on the absorption of the  $\alpha$ -rays of uranium by thin sheets of aluminum indicate a range of 3.3 cm. A very plausible explanation of the lack of agreement of the experimental value with that calculated, 6.65 cm., lies in the supposition of a new  $\alpha$ -ray product of uranium, with a range of about 3.2 cm. This excess of  $\alpha$ -ray activity is not to be accounted for to any appreciable extent, by the  $\alpha$ -ray activity of UX, since the latter activity constitutes, at most, but a very

<sup>1</sup> Phil. Mag., 11, 754 (1906).

small fraction of the total activity of uranium. The new product may be either a very rapidly changing one<sup>1</sup> between U and UX; or it may be inseparable<sup>2</sup> from uranium just as radiothorium is from thorium<sup>3</sup>. Of course there remains the possibility as we at first<sup>1</sup> also suggested that two  $\alpha$ -particles are produced directly by the disintegration of each atom of uranium.

It is very probable that altogether there are three  $\alpha$ -particles expelled during the several changes accompanying the transformation of an atom of uranium into an atom of radium. If the final product of the uranium--radium series be lead<sup>4</sup>; 8  $\alpha$ -particles of average atomic weight 3.95 are lost by an atom of uranium (atomic weight 238.5) in changing into one of lead (atomic weight 206.9). Of these 8 particles radium, and its products give off 5; leaving 3 for the change of uranium into radium. On this basis, the atomic weight of radium should be 226.65. In a paper just published<sup>5</sup> Mme. Curie shows her earlier value 225 for this constant to be too low, and gives instead 226.45; which is in good accord with the calculated value. The supposed inactive product, of very long period, between uranium and radium may be the third  $\alpha$ -ray product of uranium, which may emit  $\alpha$ -particles with less than the critical velocity necessary to produce ionization.

### Summary.

1. New determinations of the specific activity of uranium minerals were made under conditions so chosen that the  $\alpha$ -rays of the active films all reach the limits of their ranges in the air of the ionization chamber. The observed activity has been corrected for the deficiency due to loss of emanation. The specific activity of uranium in minerals is 3616, while the corresponding constant for pure uranium compounds is 796. For equal uranium content, minerals, free from thorium are  $3616/796 = 4.54$  times as active as pure compounds.

2. The activity of uranium is 1.87 times that of equilibrium amount of radium (alone).

3. The range of uranium is about 3.4 cm. There is probably a new uranium product which gives  $\alpha$ -rays with a range of 3.25 cm.

4. To explain the supposed genetic relationship of actinium to uranium, it is suggested that UX may disintegrate in two ways; the first giving  $\beta$ -

<sup>1</sup> Meyer and Schweidler.—Wien. Ber., Dec. 1904, think uranium may produce an emanation of very short period.

<sup>2</sup> Levin, Physik. Z., 7, 692 (1906), has attempted, in vain, to separate from uranium any radioactive substance other than UX.

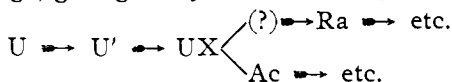
<sup>3</sup> See following paper.

<sup>4</sup> Phys. Rev., Jan. 1907.

<sup>5</sup> Boltwood, Am. J. Sci., 20, 253 (1905); 23, 77 (1907).

<sup>6</sup> Chem. News., 96, 127 (1907).

rays and the inactive product which finally produces radium; the second, much slower change, giving  $\alpha$ -rays and actinium, thus:



Oct. 12, 1907.

[CONTRIBUTION FROM KENT CHEMICAL LABORATORY, UNIVERSITY OF CHICAGO].  
**THE SPECIFIC RADIOACTIVITY OF THORIUM AND THE VARIATION OF THE ACTIVITY WITH CHEMICAL TREATMENT AND WITH TIME.**

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In our first paper<sup>1</sup> on the radioactivity of thorium compounds we showed that, in the case of minerals, when the activity due to uranium was deducted, the remaining activity was strictly proportional to the percentage of thorium present. The following table, taken from the earlier paper, summarizes the results obtained:

TABLE I.

No.	Name	% Th	% U	$k_m$	$k_U$	$k_{Th}$	$\frac{k_{Th}}{P_{Th}}$
1	Orangite. ....	43.1	7.76	649	255	394	914
2	Thorite .....	46.6	6.26	664	205	459	985
3	Monazite. ....	5.27	0.33	60.2	10.9	49.3	935
4	" .....	15.18	0.46	164	15	149	982
5	" .....	2.72	0.12	29.8	4.0	25.8	950

Mean = 953

The symbol  $k_m$ , represents the specific activity of the mineral, in terms of the activity of 1 sq. cm. of a thick film of  $U_3O_8$  as unity;  $k_U$  is the activity due to uranium;  $k_{Th} = k_m - k_U$ .  $P_{Th}$  is the weight of thorium per gram of mineral.

Results similar to these were published by Boltwood<sup>2</sup> in the same number of the Am. J. Sci. as that in which our paper appeared. Boltwood's results were expressed in arbitrary units, but showed just as clearly as do those of Table I, the constant specific activity of thorium in minerals. A third simultaneous paper, in the same journal by Dandourian<sup>3</sup> contained similar conclusions reached as a result of a different method of investigation; which consisted in the measurement of the intensity of the excited activity obtained from the thorium emanation given off by solutions of minerals. These results were also expressed in arbitrary units.

In the first paper we stated:

"All of the thorium activity measurements, as well as those of uran-

<sup>1</sup> Am. J. Sci., 21, 433 (1906).

<sup>2</sup> Ibid., 21, 409 (1906).

<sup>3</sup> Ibid., 21, 427 (1906).