

THE RELATION BETWEEN THE ALPHA-RAY ACTIVITIES AND RANGES OF RADIUM AND ITS SHORT-LIVED PRODUCTS.

BY HERBERT N. MCCOY AND EDWIN D. LEMAN.

IN a recent paper¹ it has been shown that the ratio between the α -ray activity of radioactinium and that of its products is in good agreement with the value calculated from the equation $I = kR$, where I is the ionization current in air due to a single α -particle, R is the range of the α -particle, and k is a constant for α -particles of all ranges.² McCoy and Viol³ had previously shown that the relative α -ray activities in the thorium series, as calculated from the same equation, are in good accord with the experimental values. In this paper we wish to show that the relative activities of radium and its short-lived products are also in good agreement with those calculated from the known ranges.

The latest available data on the radium series⁴ which we shall use in this paper are given in Table I.

TABLE I.

The Radium Series.				
	Symbols.	Period.	Rays.	Ranges, Cm. at 15° C.
Radium.....	Ra	1730 years	α, β	3.30
Emanation.....	Em	3.85 days	α	4.16
Radium A.....	RaA	3.00 min.	α	4.75
Radium B.....	RaB	26.7 "	β	
Radium C ₁	RaC ₁	19.5 "	$\alpha(?)$, β	
Radium C ₂	RaC ₂	1.4 "	β	
Radium C'.....	RaC'	10 ⁻⁶ sec. (?)	α	6.94

The accepted scheme of disintegration in this series, according to Fajans⁵ is as follows:

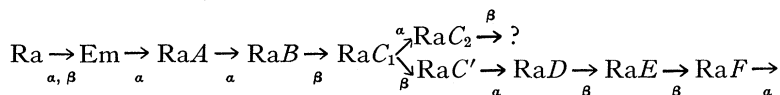
¹ McCoy and Leman, *PHYS. REV.*, 4, 409 (1914).

² Geiger, *Proc. Royal Soc., A*, 82, 486 (1909); *A*, 83, 505 (1910). Taylor, *Phil. Mag.*, 21, 571 (1911); 26, 402 (1913).

³ McCoy, *PHYS. REV.*, 1, 393 (1913).

⁴ Kolowrat, *Le Radium*, 11, 1 (1914).

⁵ *Phys. Zeit.*, 13, 699 (1912).



According to this view, the product formerly called radium *C* is complex: *C*₁, which is the product of *B*, disintegrates in two ways; the first (principal) with the production of a β -particle giving *C'*; the second (subordinate) with the expulsion of an α -particle, giving *C*₂. The product *C'* also gives α -rays, and of every 10,000 α -particles produced by the *C* components, all but three are due to the change of *C'* into *D*, the balance being due to the change of *C*₁ into *C*₂.¹ If we assume that these conclusions are essentially correct, we see that the effect on the activity caused by a minute fraction of the α -particles of radium *C* having a different range from those of the main fraction, would be negligibly small. We shall, therefore, neglect the complexity of *C* in the discussions in this paper, merely pointing out that the present work furnishes no data regarding this important question.

The method of determining the relative α -ray activities of Ra and its short-lived products consisted in preparing a Ra-BaSO₄ film free from all of the subsequent products of Ra, and measuring its initial and final activities. The activities of such films increased for about six weeks after the preparation of the film and then remained practically constant during several months of observation, the constant activity in each case being taken as the final. The Ra-BaSO₄ film, free from its subsequent products, was prepared in the following manner: One c.c. of a solution of pure radium chloride in dilute hydrochloric acid was diluted to 10 c.c.; the solution was heated to its boiling point and a current of air bubbled through it for thirty minutes, the solution being kept near its boiling point and a few c.c. of water being added from time to time to replace that lost by evaporation. This procedure kept the solution free from emanation, and allowed RaA to decay practically completely. A few drops of lead acetate solution were then added and hydrogen sulphide passed in. The lead sulphide, which was precipitated, removed *B*, *C*, *D*, *E*, and *F*. The precipitate was rapidly filtered off, and a current of air again bubbled through the filtrate, for ten minutes, keeping it near its boiling point and the volume constant. Lead sulphide was again precipitated in the solution, the precipitate filtered off, and the filtrate again treated in the manner just described, making a total of three precipitations of lead sulphide, and removing practically every trace of the products of radium. The various operations to this point had taken about fifty minutes. The filtrate from the last treatment

¹ Fajans, loc. cit.

was put into a 15 c.c. centrifuge tube, one c.c. of $N/100$ barium chloride solution added, then a few drops of dilute sulphuric acid. The solution was well shaken, and then centrifuged. The supernatant solution was decanted from the precipitate, and the latter was well washed, by decantation, with water acidified with a few drops of HCl , and finally with alcohol. A small portion of this precipitate was spread as uniformly as possible, with the aid of a glass rod and a little alcohol, over a flat polished brass plate, about 7 cm. in diameter. When the alcohol had evaporated, the less firmly adhering particles were brushed off, care being taken to guard the edges and back of the plate from radioactive contamination. The films so prepared were so thin as to be almost invisible; the differential absorption of α rays in such films was therefore negligibly small.

The time of precipitation of the sulphate was taken as zero time, as at this moment the precipitate was free from all of the subsequent products of radium. That the above method completely freed the radium from all of its active products was shown both by control experiments¹ and by the fact that the activity increased at a regular rate from the start. About thirty minutes elapsed between the time when the sulphates were precipitated and the time of making the first measurements. The activity measurements were made in a gold-leaf electroscope as previously described,² the active films being placed 7 cm. below the charged electrode, thus allowing all rays to reach their full ranges. Sufficient potential (about 600 volts) was used to insure practically complete saturation currents for the weak ionization produced. All activities were measured in comparison with a standard film of uranium oxide, the activities given being in terms of this standard. All measurements were made with the greatest care, and corrections were made for the accurately determined natural leak, which in every case was less than one per cent. of the standard. The initial activity of each film was less than, the final activity greater than, the standard. The activity increased nearly linearly for the first six hours so that by a small extrapolation the activity at time zero could be determined with a high degree of accuracy. The activities in terms of the standard are given in Table II. for two films.

TABLE II.
The Initial and Final Activities of Radium (Uncorrected).

Film.	Initial Activity I_0 .	Final Activity I .	Ratio Uncorrected.
1	0.858	4.325	5.043
2	0.558	2.832	5.075

¹ Compare the work of McCoy and Viol, loc. cit., on the separation of Th-B-C-D from ThX.

² McCoy and Ashman, Am. Jour. Sci., 26, 521 (1908).

But these results are subject to three corrections, viz.: the activity due to the β -rays, the escape of emanation from the film either by diffusion or by recoil, and the loss of other products by recoil.

The activity due to β -rays was determined by placing consecutive layers of aluminium foil over the Ra-BaSO₄ film, the first layer being of sufficient thickness to absorb all α -rays, and measuring the activity with each additional layer in the same electroscope as was used for the α -ray measurements. By plotting the activities against the thickness of the covering, and extrapolating to the axis of the activities, the β -ray activity could be ascertained with a fair degree of accuracy. This activity must be subtracted from the final activity of the film.

Since Kolowrat has shown that¹ the β -ray activity of radium free from its products is only 2 per cent. of the β -ray activity of radium in equilibrium with its short-lived products, and since the β -ray activity of the latter as measured in our electroscope was only about 0.5 per cent. of the α -ray activity, it follows that it is not necessary to apply any correction for β -ray activity of the radium itself.

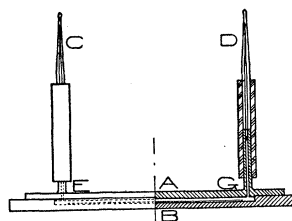


Fig. 1.

The amount of emanation lost by the film was determined by the use of a circular brass box as shown in Fig. 1. The cover, *A*, fitted with two small bore tubes *E* and *G*, was ground so as to fit very tightly over the lower compartment *B*. In the lower compartment was a recess about 0.2 cm. in depth and 8 cm. in diameter. Small bore glass tubes drawn out at one end, *C* and *D*, were attached to the tubes *E* and *G* by means of short pieces of thick-walled rubber tubing. The Ra-BaSO₄ film was placed in *B*, the cover *A* fitted on, and all joints sealed with wax. The ends of *C* and *D* were then sealed by fusion. After an interval of 40 days or more, the emanation which had accumulated was drawn into an emanation electroscope by a stream of air, care being taken to prevent any loss of the accumulated emanation and also to prevent any diminution of pressure within the box which might withdraw some of the Em from the film. The rate of discharge of the electroscope was measured, after the emanation had stood in it for three hours.

In order to find what fraction this quantity of Em was of the equilibrium quantity in the Ra-BaSO₄ film, two additional determinations were necessary. One c.c. of the original radium solution was diluted and then freed from subsequent active products as described in a preceding paragraph. By means of a small weight-pipette, an accurately

¹ Le Radium, 7, 269 (1910).

weighed portion of this solution was taken and uniformly distributed over the surface of a flat platinum plate, and evaporated to dryness. The activity of the residue, which was entirely invisible, was measured within twenty minutes, and the measurements extended over a short interval in order to be able to get the initial activity by extrapolation. To find the activity of the emanation from this quantity of radium, a known portion of the solution in the weight-pipette was run into a small round-bottom flask containing about 10 c.c. of dilute hydrochloric acid. The flask was well stopped with a two-hole rubber stopper fitted with delivering tubes, one of which extended into the solution. A current of air was bubbled through the solution for 30 minutes to drive off the accumulated emanation, and the flask then sealed by fusing the ends of the delivery tubes. After an interval of a few days, the emanation was drawn into the emanation electroscope, by allowing air to bubble through the solution which was heated near its boiling point. After three hours, the rate of discharge of the electroscope was measured.

The corrections for loss of emanation by the film were made as follows:

Initial activity of film No. 1	0.858
Initial activity of Ra on the platinum plate660

The radium on the platinum plate was obtained from 1.4942 gm. of solution. 1.5738 g. of the same solution gave after three days, 21.5 hours, a quantity of Em which discharged the electroscope in 46.45 seconds. Since for three days 21.5 hours, $1 - e^{-\lambda t} = 0.5036$, the equilibrium amount of Em from the solution would have discharged the electroscope in $0.5036 \times 46.45 = 23.40$ sec. Therefore the equilibrium amount of Em from 1.4942 gm. of this solution would discharge the electroscope in 24.64 sec. That is, a radium film, free from all subsequent products of radium, having an initial α -ray activity of 0.660 produces an equilibrium quantity of emanation which discharges the emanation electroscope in 24.64 sec. It then follows that the equilibrium quantity of emanation from film No. 1, which has an initial activity of 0.858 would discharge the emanation electroscope in 18.97 sec. The emanation which had escaped from film No. 1, when the latter had been sealed up 40 days or more, discharged the emanation electroscope in 1,748 seconds, therefore 1.08 per cent. of the equilibrium quantity of Em escaped from the film. This means that the quantity of emanation and the short-lived products in the film is 1.08 per cent. too low. In this film, the activity of the subsequent products of Ra is $4.297 - 0.858 = 3.439$, which value is 1.08 per cent. too low, hence the true activity of the products, if no Em had escaped, would be 3.476.

To find the loss of activity due to recoil, a polished brass plate 7 cm.

in diameter was placed 1 mm. above and completely insulated from film No. 1. A potential of 110 volts was maintained for six weeks, the upper plate being kept negatively charged. The activity of the plate was measured as quickly as possible after removing the potential, four minutes elapsing from the time the potential was removed to the mean time of making the measurements. The activity of the active matter which collected on the plate was 0.0374. Assuming that the active matter on the plate at the instant the potential was removed was RaA, RaB, and RaC in equilibrium, the activity of the matter on this plate four minutes after the potential is removed is approximately 70 per cent. of its initial value.¹ Therefore the initial activity of the matter on the plate was 0.0534. To find what quantity of the matter on the plate is due to active deposit from escaped emanation and what quantity is due to direct recoil, it is necessary to recall that the emanation lost by this film was 1.08 per cent. of the equilibrium quantity, or the activity of the products, Em, RaA and RaC, lost was 0.037. Assuming that the activities are proportional to the $2/3$ powers of the ranges, the fraction of the activity due to RaA + RaC is 65 per cent. of the total activity due to Em + RaA + RaC, or 0.0241 and that due to recoil is $.0534 - .0241 = .0293$.²

Table III. gives the results of two determinations. The final activity is corrected for β -ray activity, loss of emanation, and loss by recoil, these corrections being determined separately for each film:

TABLE III.

The Initial and Final Activities of Ra (Corrected).

Film.	Initial Activity I_0 .	Final Activity I .	$\frac{I_0}{I}$.
1	0.858	4.363	5.085
2	0.558	2.863	5.133
			Mean . . 5.109

¹ Rutherford, Radioactive Substances and their Radiations (1913), page 491.

² It must be pointed out that this value is calculated on the assumption that at the instant the potential is removed, the active matter on the plate is RaA + RaB + RaC in equilibrium, and there is a decided drop in activity before the first measurement is made, due to the rapid decay of RaA. In the case of the active matter due to active deposit we do have this equilibrium, but in the case of recoil atoms, there is in all probability an excess of RaC atoms, over the equilibrium number of RaA atoms. This would mean that the percentage of the activity which decayed during the interval between the time the potential was removed and the time of measurement would not be so great as that calculated upon complete equilibrium, and the value of the loss by recoil would be even less than that calculated above. However, the loss by recoil is so small, that the value calculated above is certainly correct within experimental error, and it represents the maximum value.

In the series Ra-Em-RaA-RaC it is very probable that each member when present in equilibrium amount produces the same number of α -particles per unit time, if this is so, the equation

$$I = kR^{\frac{2}{3}}$$

leads to the theoretical results shown in Table IV.

TABLE IV.

	Ranges at 15°.	$R\%$.	Relative Activities.	
			Calculated.	Found.
Ra.	3.30	2.217	1.00	[1.00]
Em.	4.16	2.586	1.17	4.11
RaA.	4.75	2.826	1.28	
RaC.	6.94	3.639	1.64	

The only previous determination of the relative activities of radium and its products was made by Boltwood.¹ In these experiments the films were made by the evaporation of a chloride solution. Since such films gave off very large fractions of the emanation produced (in one case as high as 65 per cent.) it is not surprising that the results obtained were not very accurate. Instead of a ratio of 4.11 as found by us (last column, Table IV.) Boltwood found 4.65. For several years this result was looked upon as satisfactory,² since it was in good agreement with that calculated upon the assumption that the number of ions produced by an α -particle is proportional to the first power of its range³ instead of the two-thirds power.

The good agreement of our results with the theoretical value as calcu-

TABLE V.

Relative Alpha-Ray Activities.

Ratios Measured.	Found.	Calculated.
Thorium X and products to radiothorium ¹	3.66	3.60
Thorium C ₁ and C ₂ to radiothorium ¹	1.54	1.56
Actinium X and products to radioactinium ²	4.67	4.63
Radium emanation and products to radium ³	4.11	4.09

¹ McCoy and Viol, loc. cit.

² McCoy and Leman, loc. cit.

³ McCoy and Leman, this paper.

lated by the equation $I = kR^{\frac{2}{3}}$, the correctness of which may be considered as already established, shows that only those members which we

¹ Phys. Zeit., 7, 489 (1906); Le Rad., 3, 170 (1906).

² Rutherford, Radioactive Substances, p. 447 (1913).

³ Boltwood, loc. cit.

have considered play significant parts in the α -ray activity of the radium series, and that the accepted ranges are at least approximately correct.

In Table V the results of the recent investigations carried out in this laboratory along analogous lines are brought together.

The values headed "Found" are the relative observed α -ray activities of the indicated substances. The calculated values were obtained by means of the equation $I = kR^{\frac{2}{3}}$, according to which the number of ions produced by an α -particle is proportional to the $2/3$ power of its range.

KENT CHEMICAL LABORATORY,
UNIVERSITY OF CHICAGO,
March 3, 1915.