

THE VARIATION OF THE ALPHA-RAY IONIZATION OF  
RADIOACTIVE SOLIDS WITH THE THICKNESS OF  
THE LAYER.

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THE subject indicated by the title of this paper was first studied by Rutherford and McClung,<sup>1</sup> who assumed an exponential absorption law. Later the writer<sup>2</sup> studied the matter extensively after having devised a method of making very thin layers or films of uranium compounds of quite uniform thickness. The results were in satisfactory agreement with an exponential absorption law. After Bragg's discovery that an  $\alpha$  particle has a definite range, it became evident that there could be no true "absorption" of the  $\alpha$  rays and therefore that an exponential law could not serve as a logical explanation of the facts. Bragg,<sup>3</sup> St. Meyer and v. Schweidler<sup>4</sup> and others have studied the closely related problems of the activity of a thick or thin active layer covered with one or more thin foils.

The work of Bragg and Kleeman<sup>5</sup> and also McClung<sup>6</sup> supplemented by the more recent work of Geiger<sup>7</sup> and Taylor<sup>8</sup> shows the true shape of the ionization curve. Geiger found that the ionization at any point in the path of a particle is inversely proportional to the cube-root of the remainder of its range. This leads to the conclusion<sup>9</sup> that the total ionization of an  $\alpha$  particle of range  $R$  is proportional to  $R^{\frac{2}{3}}$ . Although it is very probable that this empirical law is not absolutely exact it is at least a close approximation and as such is very useful in several ways. In a paper just published with C. H. Viol,<sup>10</sup> it was shown that the relative activities of the  $\alpha$ -ray products of thorium are very closely proportional to the  $\frac{2}{3}$  powers of the ranges of their respective  $\alpha$ -rays. I have also found a similar result in a still more recent, unpublished, study of radium and its products.

<sup>1</sup> Phil. Trans., A, 196, 25 (1901).

<sup>2</sup> Jour. Amer. Chem. Soc., 27, 391 (1905).

<sup>3</sup> Phil. Mag., 11, 754 (1906).

<sup>4</sup> Wien Sitzungsberichte, 115, II. a, 736 (1906).

<sup>5</sup> Phil. Mag., 8, 726 (1904); 10, 318 (1905).

<sup>6</sup> *Ibid.*, 11, 131 (1906).

<sup>7</sup> Proc. Roy. Soc., A, 82, 486 (1909).

<sup>8</sup> Amer. Journ. Sci., 28, 357 (1909); Phil. Mag., 21, 571 (1911); 23, 670 (1912).

<sup>9</sup> Taylor, Phil. Mag., 21, 571 (1911).

<sup>10</sup> Phil. Mag., 25, 333 (1913).

If we assume that the total number of ions produced in air by any  $\alpha$  particle is proportional to the  $2/3$  power of the distance it travels in air after leaving a radioactive layer, we may readily get an expression for the activity of such a layer as a function of its thickness. Let  $n$  = the number of  $\alpha$  particles per second from 1 g. of the substance. The number shot upward from 1 sq. cm. of a layer of thickness  $dl = \frac{1}{2} n \rho dl$ , where  $\rho$  is the density. These go equally to every element of a circumscribed hemisphere of surface  $2\pi a^2$  ( $a$  = radius). Those rays which emerge between the angles  $\theta$  and  $\theta + d\theta$  to a normal strike a zone of area  $2\pi a^2 \sin \theta d\theta$  and therefore constitute the fraction  $\sin \theta d\theta$  of the rays shot upward; so that the number of rays reaching the zone is  $\frac{1}{2} n \rho \sin \theta d\theta dl$ . Now suppose the current due to a single  $\alpha$  ray is  $Cr^{\frac{2}{3}}$  where  $C$  is a factor which is constant for the  $\alpha$  rays of all substances and  $r$  is the effective air range. If  $R$  is the range when there is no retardation in the solid, it is readily seen that  $(R - kl/\cos \theta) = r$  is the effective range of the emerging ray, if  $l$  is the depth from which the  $\alpha$  particle comes and  $k$  is the ratio of  $R$  to the "range"  $l_0$  of the  $\alpha$  particle in the solid; that is  $k = R/l_0$ . The current due to rays between  $\theta$  and  $\theta + d\theta$  from an infinitesimal thickness  $dl$  at a depth  $l$  from the upper surface is  $\frac{1}{2} n \rho C (R - kl/\cos \theta)^{\frac{2}{3}} \sin \theta d\theta dl$  and for the whole of the rays is

$$dA = \frac{1}{2} n \rho C \int_0^\theta \left( R - \frac{kl}{\cos \theta} \right)^{\frac{2}{3}} \sin \theta d\theta dl,$$

where the upper limit of  $\theta = \cos^{-1} kl/R$ ; since if  $\theta > \cos^{-1} kl/R$  the  $\alpha$  rays from the lower surface do not get out of the film. If  $l_0$  is the "range" in the solid and  $l/l_0 = x$ ;  $x$  is the thickness of a film in terms of the thickness just necessary to give maximum activity. Since  $k = R/l_0$ ;  $kl/R = l/l_0 = x$  and therefore

$$dA = K dx \int_0^{\cos^{-1} x} \left( 1 - \frac{x}{\cos \theta} \right)^{\frac{2}{3}} \sin \theta d\theta,$$

where  $K = \frac{1}{2} n \rho C R^{\frac{2}{3}} l_0$ , a constant for a given substance. Therefore the ionization current of a film of thickness  $x$  is

$$A = K \int_0^x dx \int_0^{\cos^{-1} x} \left( 1 - \frac{x}{\cos \theta} \right)^{\frac{2}{3}} \sin \theta d\theta.$$

If the indicated integral of function  $\theta$  is written  $f(x)$  then

$$A = K \int_0^x f(x) dx.$$

As the  $\theta$  integration gave a very complex expression which presented

difficulties for the  $x$  integration, the ordinary method was abandoned and the integrations accomplished without difficulty, by graphic methods. The results are shown in Table I., in which  $A$  is expressed in terms of the maximum activity as unity, the numbers in the last column being proportional to those in the third column.

TABLE I.

$x$	$f(x)$	$\int f(x)dx$	$A$
0.0	57.30	0.00	0.000
0.1	41.58	4.87	0.284
0.2	31.85	8.52	0.497
0.3	24.06	11.31	0.659
0.4	17.67	13.39	0.780
0.5	12.61	14.90	0.869
0.6	8.44	15.95	0.930
0.7	5.01	16.59	0.967
0.8	2.44	16.96	0.989
0.9	0.76	17.12	0.998
1.0	0.00	17.16	1.000

The graph of  $A$  as a function of  $x$  is practically identical with an exponential curve up to about  $A = 0.5$ ; while for larger values of  $x$  it lies only 1 to 3 per cent. higher. Since my earlier work on uranium compounds and also that of Goettsch<sup>1</sup> had shown agreement with the exponential law within the experimental error of less than 1 per cent., excepting for films of nearly maximum activity where the deviation was a little greater, it follows at once that the facts are also in good agreement with the formula developed above. However the earlier experiments did not conform strictly to the conditions assumed in the derivation of the formula in that the films were made in shallow tins, having rims 0.8 cm. high instead of on flat plates; also, the uranium used contained UX the  $\beta$  rays of which produced a measurable part of the observed activity and finally uranium itself gives two sets of  $\alpha$  rays of different ranges.<sup>2</sup>

<sup>1</sup>Jour. Amer. Chem. Soc., 28, 1540, 1906.

<sup>2</sup>The suggestion that uranium gives two sets of  $\alpha$  rays has occasionally been credited to Boltwood, Amer. Jour. Sci., 25, 298 (1908). I should like to point out that the first announcement of this idea, as far as I can find, was that by Dr. Ross and myself at the Chicago meeting of the Amer. Phys. Soc., Dec. 1, 1906. The abstract of this paper dated November 23, 1906, appeared in the PHYS. REV., January, 1907. In this paper as a result of our measurements of the relative activities of uranium and radium in minerals we concluded that "Since uranium is about twice as active as the equilibrium amount of radium the former must produce two  $\alpha$  particles for every one produced by the equilibrium amount of the latter. This indicates that each atom of uranium produces upon disintegration two alpha particles or that there is an alpha-ray product not yet isolated between uranium and uranium X." Dr. Boltwood,

The first two conditions could easily be corrected experimentally, while the effect of two sets of  $\alpha$  rays could be treated theoretically.

For an infinitely thin film  $f(x) = 57.30$ ; while for  $x = 1$ ,  $\int f(x)dx = 17.16$ ; Table I. The ratio  $\frac{17.16}{57.30} = 0.2994$  is the activity of a film of

just sufficient thickness,  $l_0$ , to have maximum activity, in terms of the activity of the same mass of material spread into an infinitely thin film on a flat surface. This ratio may be taken practically as 0.3. The activity of a thick film of a uranium compound, say  $U_3O_8$ , is the sum of the partial activities of the two sets of  $\alpha$  rays. If we call the maximum partial activities of 1 sq. cm.  $A_1$  and  $A_2$  respectively, we may easily find theoretically the relative magnitudes of  $A_1$  and  $A_2$ . Let  $W_1$  be the weight of 1 sq. cm. of a film just thick enough to have maximum activity for the  $U_1$  rays (those of uranium itself) and  $W_2$  be the weight for  $U_2$  rays. Let  $a_1$  be the activity due to  $U_1$  rays of 1 g. in an infinitely thin film on a flat plate and  $a_2$  be the activity for the  $U_2$  rays. Then  $A_1 = 0.3W_1a_1$  and  $A_2 = 0.3W_2a_2$  but  $W_1/W_2 = R_1/R_2$  where  $R_1$  and  $R_2$  are the respective ranges and

$$\frac{a_1}{a_2} = \left(\frac{R_1}{R_2}\right)^{\frac{3}{2}}$$

Therefore

$$\frac{A_1}{A_2} = \left(\frac{R_1}{R_2}\right)^{\frac{3}{2}}$$

For uranium Geiger<sup>1</sup> has found  $R_1 = 2.37$  and  $R_2 = 2.75$  at  $0^\circ$  and 760 mm. Therefore  $A_1 = 0.781 A_2$  and  $a_1 = 0.906a_2$ ; also

$$A_1 + A_2 = 0.3 \times 0.934(a_1 + a_2)W_2.$$

If a film is thick enough to have maximum activity for the more penetrating  $U_2$  rays the activity due to the rays of uranium itself ( $U_1$ ) should be 0.781 of the activity due to  $U_2$ . Maximum activity for  $U_1$  rays would be reached for a thickness  $R_1/R_2 = 0.862$  as great as is required for the  $U_2$  rays. By plotting a curve with  $x$  and  $A$  as coördinates and adding

whose first communication on this subject is in a letter dated December 17, 1906, to Nature, January 3, 1907, wrote: "It will be noted in the above that the activity of uranium is about twice that of the radium present, which is in good agreement with the conclusion of Moore and Schlundt that there are two alpha ray changes in uranium if it is assumed that the average range of the two uranium particles is about 3.5 cm." Dr. Boltwood's conclusion was based, therefore, on the same facts (obtained in independent experiments) as our own. While it is true that Moore and Schlundt did think that UX gives alpha rays, thus suggesting a second alpha-ray change in uranium; this was disproved later and so has nothing to do with the matter in question. See also McCoy and Ross, Jour. Amer. Chem. Soc., 29, 1708 (1907).

<sup>1</sup> Phil. Mag., 24, 653 (1912).

to it a curve of the same form, but having its maximum ordinate 0.781 of that of the first curve while its maximum abscissa is 0.862 we get the theoretical curve for the total activity of a film of any uranium compound as a function of its thickness, where the thickness is expressed as a fraction of that required for maximum activity of the  $U_2$  rays. (See Fig. 1.) Table II. gives the total activities for various values of  $x$ . It is seen by comparison with Table I. that the total activities thus found are, for each value of  $x$ , a little greater than for a substance giving but one set of  $\alpha$  rays.

TABLE II.

$x$	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0
Activity	0.300	0.522	0.686	0.810	0.894	0.945	0.977	0.993	0.999	1.000

To test the accuracy of these deductions as carefully as possible new experiments were made with films of  $U_3O_8$ . Pure uranium nitrate was completely freed from UX and also any possible trace of radium by means of barium sulphate in the usual way. The nitrate was decomposed by heat and the oxide heated to constant weight at  $700^\circ$  in an

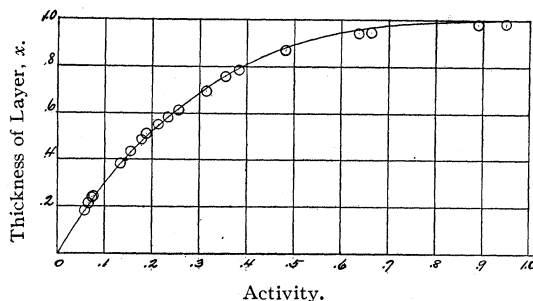


Fig. 1.

Theoretical curve with experimentally determined points.

electric muffle.<sup>1</sup> The product so obtained was pure  $U_3O_8$ . This material was made into films on flat circular brass or copper plates in the manner previously described,<sup>2</sup> alcohol being used as the suspending liquid. The films were dried at  $105^\circ$ ; those that showed any unevenness or imperfection were discarded. The activity measurements were made in the gold-leaf electroscope described by McCoy and Ashman. The errors of measurement were not over 0.2 to 0.3 per cent.

Two series of films were made; the first on copper plates 7.00 cm. in diameter; the second on brass plates 6.914 cm. in diameter. The results

<sup>1</sup> McCoy and Ashman, Amer. Jour. Sci., 26, 523 (1908).

<sup>2</sup> McCoy and Ross, Jour. Amer. Chem. Soc., 29, 1699, 1907.

are given in Table III. and plotted in Fig. 1, where the points are seen to fall very close to the theoretical curve.

TABLE III.

First Series.			Second Series.		
Weight in Mg. per. Sq. Cm.	Observed Activity.	Calculated Activity.	Weight in Mg. per Sq. Cm.	Observed Activity.	Calculated Activity.
0.544	0.179	0.165	0.775	0.240	0.224
0.683	0.217	0.200	0.812	0.243	0.232
1.385	0.392	0.375	1.878	0.485	0.478
1.637	0.437	0.430	1.950	0.498	0.491
1.996	0.512	0.500	2.291	0.569	0.552
2.258	0.551	0.545	3.780	0.766	0.760
2.481	0.590	0.594	5.119	0.867	0.879
2.715	0.612	0.616	9.458	0.980	0.997
3.339	0.693	0.706	10.09	0.985	0.999
3.768	0.759	0.758	12.83	0.991	1.000
4.074	0.783	0.794	13.57	1.000	1.000
6.764	0.941	0.959	16.37	1.000	1.000
7.033	0.946	0.966			
19.87	1.000	1.000			

In order to calculate the theoretical activities it is first necessary to find a factor which converts the weight into the fraction which this weight is of that necessary to give maximum activity. This factor will be a constant for all films of  $U_3O_8$ ; it may be found from any one determination or the mean of several. This factor proved to be 94; so that  $x = 94 W$  where  $W$  is the weight in g. per sq. cm. The values given as "calculated activity" are those corresponding to the value of  $x = 94 W$  indicated by the theoretical curve (Fig. 1). We see that the agreement between facts and theory is quite close; not, perhaps, within the experimental error. But this is not to be expected, as some of the assumptions made in deriving the formula are not absolutely exact. Thus, the assumption that the factor  $k$  is a constant for a given substance is not strictly in accord with the fact that the stopping power of a metallic foil is somewhat dependent upon the velocity of the  $\alpha$  particle.<sup>1</sup> The error caused by taking  $k$  constant is not large and probably would not greatly change the theoretical curve.

By means of an equation given in a foregoing paragraph it is easy to get a second test of the accuracy of the theoretical assumptions made in this paper. It was shown that

$$A_1 + A_2 = 0.3 \times 0.934 (a_1 + a_2) W_2.$$

<sup>1</sup> Bragg, Phil. Mag., 13, 507 (1907); Taylor, Amer. Jour. Sci., 28, 357 (1909); Phil. Mag., 18, 604 (1909).

If we take the  $\alpha$ -ray activity of 1 sq. cm. of a thick film of  $U_3O_8$  as the unit of activity, then  $A_1 + A_2 = 1$  and

$$a_1 + a_2 = \frac{1}{0.3 \times 0.934 W_2}$$

$a_1 + a_2$  is the activity that 1 g. of  $U_3O_8$  would have in the form of an infinitely thin film on a flat surface. The value of  $a_1 + a_2$  is readily found from the weight and observed activity of films as given in Table III. The ratio of activity to weight increases with the thinness of the film and reaches a maximum for a vanishingly thin film. By a short and quite definite extrapolation of this ratio, as determined by the values given in Table III., it was found that  $a_1 + a_2 = 335$ .<sup>1</sup> Since  $x = 94W$  if  $x = 1$ ,  $W_2 = 1/94 = 0.01064$ , the weight per sq. cm. just necessary to give the maximum  $U_2$  activity. By substituting this value in the above equation  $a_1 + a_2 = 336$ . The close agreement between the values found in the two ways is further proof of the correctness of assumptions made in the theoretical discussion.

The "range" of the  $\alpha$  rays in  $U_3O_8$  is easily calculated from the above data. If  $\rho$ , the density of  $U_3O_8$ , is taken as  $7.31$ ,<sup>2</sup>  $l_0$ , the range of the  $U_2$  rays =  $0.01064/7.31 = 0.00145$  cm. The range of the  $U_1$  rays is  $R_1/R_2 \times 0.00145 = 0.00125$  cm. Then the constant  $k = R_2/l_0 = 1890$  for both sets of rays. By substituting for the ranges in air and  $U_3O_8$  the weights per sq. cm., which an  $\alpha$  ray can penetrate we get a constant into which a knowledge of the density of  $U_3O_8$  does not enter. The mass of a column of air 1 sq. cm. in section and 2.75 cm. in length is 0.00355 g.; while  $W_2 = 0.01064$  g. The ratio of the latter weight to the former is 3.01. This is the ratio of the weights of  $U_3O_8$  and air which produce the same retardation of an  $\alpha$  particle; it is the same for both sets of rays of  $U_3O_8$ .

The relative "absorption" or, properly speaking, retardation of the  $\alpha$  rays in passing through matter of different kinds has been studied extensively. Bragg and Kleeman<sup>3</sup> found that the stopping power of a substance was an atomic property and for equal masses per sq. cm. nearly inversely proportional to the square-root of the atomic weight. I arrived at similar conclusions<sup>4</sup> in a somewhat different way from that

<sup>1</sup> The value, 335, is only half of the total activity, as half of the rays are absorbed in the plate on which the film is made. The total activity is therefore  $2 \times 335 = 670$ . Since  $U_3O_8$  contains 84.8 per cent. of uranium, this corresponds to an activity of 790 for 1 g. of uranium. This is exactly the value I found in the earliest determination of this constant: Jour. Amer. Chem. Soc., 27, 391 (1905); while with Dr. Ross, later, *Ibid.*, loc. cit., the value found was 796.

<sup>2</sup> Ebelman, Ann. Chim. phys. (3), 5, 189 (1842).

<sup>3</sup> Phil. Mag., 10, 318 (1905).

<sup>4</sup> Jour. Amer. Chem. Soc., 28, 1555 (1906).

of Bragg and Kleeman. The conclusions reached in the preceding paragraph may easily be compared with the results expected by the square-root law. By this law we calculate that 59.1 per cent. of the retardation that an  $\alpha$  particle experiences in passing through a layer of  $U_3O_8$  is caused by the uranium and 40.9 per cent. by the oxygen. Since 0.01064 g. of  $U_3O_8$  containing 84.8 per cent. of uranium, will completely stop a  $U_2$  particle; the same result would be produced by 0.0153 g. of uranium per sq. cm. or by 0.00400 g. of oxygen. The corresponding calculated values for nitrogen and air are 0.00374 g. and 0.00377 g. The ratio of 0.01064 to 0.00377 is 2.82; while the experimental result is 3.01. The calculated ranges in air at  $0^\circ$  and 76 cm. are 2.52 cm. and 2.92 cm. for  $U_1$  and  $U_2$  instead of 2.37 cm. and 2.75 cm. as found by Geiger. The differences are probably largely due to inexactness of the square-root law.

## SUMMARY.

1. A formula has been developed for the  $\alpha$ -ray activity of a layer of a radio-active solid as a function of its thickness. It was assumed that the air range of an  $\alpha$ -ray is diminished in its passage through the solid by an amount directly proportional to the distance it travels in the solid before emerging and that the ionization produced is proportional to the  $2/3$  power of the distance the  $\alpha$ -ray then travels in air. The theoretical curve for the rise of activity with increasing thickness of the layer is almost the same as an exponential curve, although the fundamental equations are entirely different.

2. It has been shown that the activity of a film of any  $\alpha$ -ray substance just thick enough to give maximum activity is 0.3 of the activity that the same material would have if spread on a flat surface in an infinitely thin film.

3. It has been shown that the partial activities of a thick film which gives equal numbers of  $\alpha$ -rays of ranges  $R_1$  and  $R_2$  are in the ratio  $(R_1/R_2)^{2/3}$ .

4. It has been found experimentally that the activities of films of  $U_3O_8$  were in good agreement with those calculated by theory.

5. The weights per sq. cm. of  $U_3O_8$  and of air which produce equal retardation of an  $\alpha$ -ray are as 3.01 to 1. This ratio is approximately that calculated by the law, first stated by Bragg and Kleeman, that the stopping power of matter is approximately inversely proportional to the square-root of the atomic weight of the material through which an  $\alpha$ -ray passes.

In the experimental part of this work I have been ably assisted by Mr. Edwin D. Leman, to whom I wish here to express my sincere thanks.

KENT CHEMICAL LABORATORY,  
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