[Contribution from the Rare and Precious Metals Experiment Station of the Department of the Interior, Bureau of Mines, in Coöperation with the Mackay School of Mines, University of Nevada]

CHEMICAL ACTION PRODUCED BY RADON
IV. CHARACTERISTICS OF THE ALPHA-RAY BULB AS A SOURCE OF IONIZATION[1]

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The corrections necessary to reduce the experimental efficiency factors reported in the preceding paper to the exact value, $(0.61 \pm 0.01)\ r$, (the mean effective path of α particles in a sphere of radius r) will be considered.

The construction of α-ray bulbs was first developed by Lind and Duane and has been described in detail by Lind.[2] Although very thin bulbs are desirable, there is a practical limit of thinness, dependent on the diameter of the bulb. Moreover, the α-ray bulb is not a perfect spherical shell; it has a tip, t, (Fig. 1) of relatively thick glass and a capillary connection m. Alpha particles striking either the tip or mercury in the neck (at the shoulder of the bulb) are lost. This loss should be made as small as possible, but reduction of the diameter of the neck below 0.4 mm. is not practical, for it would be difficult to maintain a perfect setting of the mercury. The column of mercury in a very small neck has a tendency to separate, throwing a droplet of mercury into the bulb, which it is impossible to reunite. The complete corrections necessary to establish the α radiation from such bulbs require rather laborious calculations for each set of dimensions. From the manipulative standpoint, bulbs 1.5 mm. in diameter with wall thickness of 0.0025 mm. are most satisfactory. Hence, we have used bulbs as nearly as possible of this size. Our calculations regarding ionization will be rigidly correct only for such α-ray bulbs.

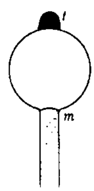

Fig. 1

1. Correction for Tip and Neck.—In a bulb of 1.5 mm. diameter, containing radon, we have assumed that RaA and RaC (in equilibrium) are deposited uniformly upon the wall, and that from any point in the bulb or upon the wall, the emission of α particles is uniform in every direction. Let a and b be the solid angles subtended from the center of the bulb by the tip and neck, respectively. The fraction of the total surface represented by the tip and neck is $(a + b)/4\pi$. The fraction of the surface which is effective in radiating is $1 - [(a + b)/4\pi]$. If $k\mu/\lambda$ is the ve-

[1] Published with the permission of the Director of the Bureau of Mines.

[2] Lind, *Am. Chem. J.*, **47**, 400 (1912). "Chemical Effects of Alpha Particles and Electrons," Chemical Catalog Co., N. Y., **1921**, pp. 76–77.

locity constant for an actual α-ray bulb, the velocity constant for a theoretically perfect bulb (without tip or neck) is $\frac{k\mu}{\lambda} \cdot \frac{1}{1 - \frac{a+b}{4\pi}}$.

2. Correction for the Radius of the α-Ray Bulb.—Let r be the radius of the reaction sphere and R be the radius of the α-ray bulb. The distance traversed by an α particle emerging normally from the α-ray bulb is[3] $r - R$. For the theoretical case of an α-ray bulb of zero radius, the distance traversed would be r. The velocity constant for a theoretical α-ray bulb (with no tip or neck and zero radius) is $\frac{k\mu}{\lambda} \cdot \frac{1}{1 - \frac{a+b}{4\pi}} \cdot \frac{\gamma}{r - R}$.

3. Calculation of the Effective Average Intensity of Ionization.—In calculating the ionization produced by any given α particle emerging from the bulb and traversing the gas phase, two factors must be considered: (1) the obliquity of passage through the wall;[4] (2) the change of ionization along the path as expressed by the Geiger curve. Evidently (2) is dependent upon (1). The ionization (Fig. 2) from (o) to (l) is represented approximately by an expression of the type,[5] $y = \frac{K}{(R - x)^{1/3}}$ where y is the intensity of ionization at any point x, and R is the range from (o) to (l). The constant K is dependent upon the gas traversed and its pressure. The end of the curve (l) to (m) is nearly a straight line and must be represented by a separate equation. Bragg[6] showed that the shape of the ionization curve is only approximately the same for all gases when the abscissas are adjusted to represent the same stopping power. Thus, Henderson's curve[7] for air is not exactly similar to the one found by Geiger for hydrogen. For the electrolytic mixture of hydrogen and oxygen used, we therefore construct a composite curve (Fig. 2) from the separate curves for hydrogen (Geiger) and air (Henderson), giving the hydrogen values double weight. The abscissas are retained as those of air at 20° and 760 mm. pressure. The ranges of α particles from Rn, RaA and RaC are taken as 4.23, 4.83 and 7.06 cm., respectively. For the pressure changes in Expts. 1 and 2 of the preceding paper the path of an α particle from the inner to the outer bulb is equivalent to an abscissa difference varying between 0.40 and 0.75 cm. A preliminary set of calculations of the ionization showed that the *average intensity of ionization* from an α-ray bulb

[3] The correction for oblique rays is taken up in Section 3.

[4] The effect of obliquity was treated graphically by Lind in ozone formation (Ref. 2). In those experiments, the reaction chamber was so large that no α particles reached the outer wall, so that Correction 2 did not arise.

[5] Geiger, *Proc. Roy. Soc.*, **83A**, 505 (1910).

[6] W. H. Bragg, "Studies in Radioactivity," **1912**, Chapters 5 and 6.

[7] Henderson, *Phil. Mag.*, **42**, 538 (1921).

is constant through this range of path. We therefore calculate the *effective average intensity of ionization* of α particles emerging from an α-ray bulb as used in these experiments as follows.

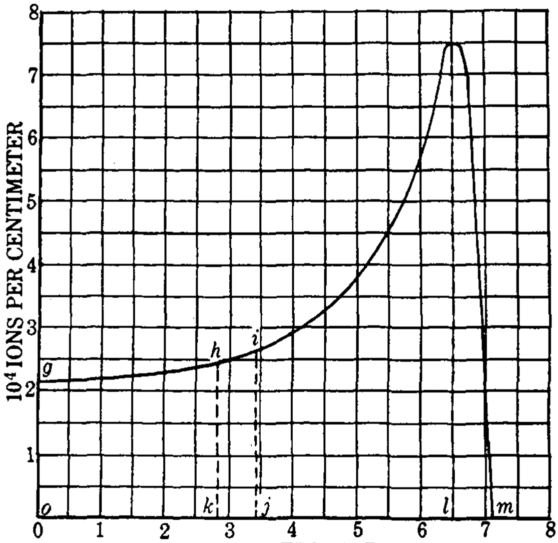

Fig. 2

Consider a point b on the inner surface of the α-ray bulb shell abc (Fig. 3). Θ is the angle between the oblique pencil of rays be and the normal pencil bd. For increasing values of Θ, the path through the glass wall increases until at 87° no α particles, even from RaC, emerge from the bulbs used. For angles of Θ greater than 90°, the pencil passes through the interior of the α-ray bulb before it reaches the wall. However, the wall thickness traversed by such pencils is the same as for the supplementary angle, due to the symmetry of a secant intercepting the walls of a spherical shell. It is therefore necessary to consider only the average effect of α particles emerging at angles less than 90°. The thickness of glass traversed for increasing values of Θ was calculated geometrically from the diameter and wall thickness. A plot was made of the thickness

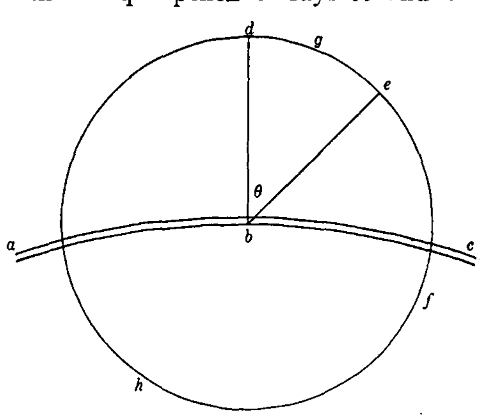

Fig. 3

as a function of Θ (Fig. 4). The lengths of path (in terms of air) cut off by the shell were listed for the following angles: 0°; 7.5; 15; 22.5; 30.0, 37.5, 45; 52.5; 60; 67.5; 71.25; 75; 76.8; 78.8; 80.63; 82.5; 84.38; 86.25; 87.2, 88.13, 90.0. They are represented (Fig. 2) by the variable *ok*. The path in the reaction vessel for each pencil is represented by *kj*; and the number of ions by the area *hijk*. The area divided by *kj* gives an average ordinate for each pencil which we shall call *intensity of ionization*.

The next step is to determine the relative number of α particles emerging in each pencil. For convenience, circumscribe a sphere of radius *bd* about the point *b* (Fig. 3). A circular section *dgefh* is cut out by a plane through the normal to the surface of the α-ray shell at *b*. Assume

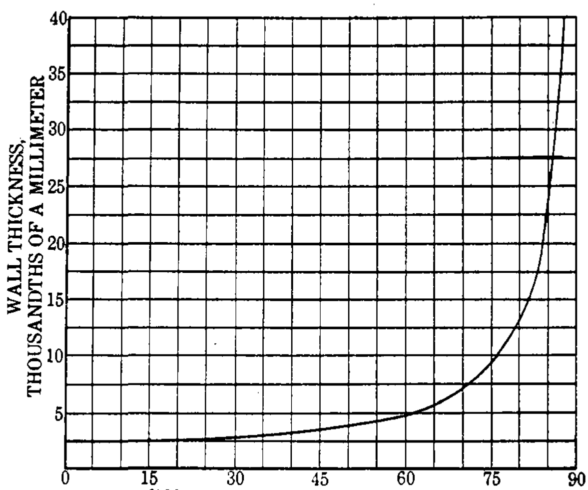

Fig. 4

that the intensity of ionization for a pencil with angle Θ_n is the average for the bundle bounded by $\Theta_n - {}^1/_2(\Theta_n - \Theta_{n-1})$ on one side and by $\Theta_n + {}^1/_2(\Theta_{n+1} - \Theta_n)$ on the other, where Θ_n is the *n*th angle. The bundle thus bounded we designate as $\Delta\Theta_n$. Further reduction (see list given above) of the size of $\Delta\Theta_n$ does not appreciably increase the accuracy of the summation that follows. If $\Delta\Theta_n$ is revolved about the normal *bd*, it cuts out a zone of revolution from the circumscribing sphere. The area of this zone divided by half the area of the circumscribing sphere represents the fraction of α particles emerging in the bundle $\Delta\Theta_n$.

An additional variable must be considered. In Section 2 a correction for the radius of the α-ray bulb was discussed. The distance traversed by an α particle from the inner to the outer bulb is also a variable and a function of Θ. As Θ increases, this distance increases. To correct for this increase in distance, the fraction of α particles in the bundle $\Delta\Theta_n$ is weighted by multiplying by the ratio of the distance at the angle Θ_n to the normal distance.

A summation of the products of the *intensity of ionization* for each bundle by the *effective fraction* of α particles emerging in the bundle, gives the *effective average intensity of ionization* of all the α particles emitted by one of the radioactive elements. These calculations were made for RaA and RaC (assumed to be on the wall of the α-ray bulb) and for Rn (gaseous distribution). For radon (gaseous distribution) the α-ray bulb was divided into eight spherical shells. The average intensity of ionization was calculated for the mean radius of each shell and multiplied by the percentage of the volume included. Since the average intensity of ionization increases more rapidly for points near the surface, outer shells were chosen to contain smaller volumes. The average for the three sets will be designated as I_A. For Expt. 1A (preceding article), $I_A = 2.659 \times 10^4 \frac{\text{ions}}{\text{cm.}}$ (referred to air at 20°, 760 mm.) and for Expt. 2A, $I_A = 2.645 \times 10^4 \frac{\text{ions}}{\text{cm.}}$.

4. Correction for Dead Arm.—As explained in the preceding paper, a dead arm containing gas which is not radiated was necessitated in mounting α-ray bulbs for Expts. 1 and 2. The quantitative effect of the dead arm upon the velocity constant is easily deduced by examining the differential equation for the rate of change in pressure. Let $-\frac{dP}{dt} = k\mu E_0 e^{-\lambda t}.P$ for the case where there is no dead arm. If a dead arm is attached the effectiveness of the α particle is not altered,[8] and hence the same number of molecules react per unit time. But the total volume of gas has been increased by the volume of the dead arm; therefore the rate of change in pressure with dead arm will be equal to $\frac{\text{vol. of bulb proper}}{\text{vol. of bulb proper} + \text{dead arm}} \times$ rate of change of pressure without dead arm. These relations are not altered in the integrated form of the velocity equation. The velocity constant in Expts. 1A and 2A would be, if there were no dead arm, equal to: $\frac{k\mu}{\lambda}$ (Expt.1) $\times \frac{\text{vol. of bulb proper} + \text{dead arm}}{\text{vol. of bulb proper}}$. The numerical substitution of values for these corrections in Expts. 1 and 2 will be deferred until the corrections for mixtures are discussed.

5. Average Intensity of Ionization when Radon is Mixed with the Gases.—A method similar to that employed in Section 3 was used to calculate the *average intensity of ionization in mixtures*. The method of obtaining average ordinates was different in that, for the case of mixtures,

[8] A few oblique α particles are probably directed into the dead arm, but they are at least compensated by those oblique α particles which are lost in the projections of the tip and neck of the α-ray bulb outside the bulb.

no part of the path of an α particle is cut off by glass before it traverses the gas phase. The method of weighting was identical with that in Section 3. Values of Θ from 0° to 180° in intervals of 7.5°, were used. Calculations were made, first, for RaA and RaC assumed to be on the wall of the reaction sphere; second, for gaseous distribution of Rn, RaA and RaC. To obtain the values for gaseous distribution, the *average intensity of ionization* was calculated for five points along the radius of the reaction sphere so placed as to be the average radius for approximately equal volumes of gas. For radon the *average intensity of ionization* for points on the wall and at the center were, respectively, 2.518×10^4 and 2.576×10^4 ions per cm. The values for the other points lay at approximately equal intervals between these two values. We therefore took the mean of the two extremes. For Radium A, the *average intensity of ionization* is less than 1% higher for gaseous distribution than for the wall, and for RaC it is the same in both cases. It is therefore unnecessary to make any assumption in regard to the distribution of RaA and RaC. The *average intensity of ionization* for the three sets of α particles, in mixtures we designate as I_B. For Expt. 1*B* (at 350 mm.), $I_B = 2.375 \times 10^4 \frac{\text{ions}}{\text{cm.}}$ (referred to air at 20° and 760 mm.); and for Expt. 2*B* (at 450 mm.), $I_B = 2.398 \times 10^4 \frac{\text{ions}}{\text{cm.}}$.

6. Recoil Atom Effect.—Lind[9] has shown that recoil atoms from Rn, RaA and RaC produce chemical action to an extent which cannot be neglected in Expts. 1*B* and 2*B*.[10]

The combined chemical effect of α rays and recoil atoms is double the α-ray effect at 116.8 mm. of electrolytic hydrogen and oxygen in a sphere of 1 cm. diameter. Assuming that the relations are similar in large bulbs (for example that in one of 2 cm. diameter the rate is doubled at a pressure of 116.8/2 mm., etc.) the relative effects can be calculated for the mixtures in Expts. 1 and 2. Evidently recoil atoms are absent when α-ray bulbs are used, which also has recently been experimentally proved.[10]

In Expt. 1*B* at a pressure of 350 mm. the α-ray effect is 91.6% of the combined effects. In Expt. 2*B*, at a pressure of 450 mm. it is 94.4%.

7. Application of Corrections.—The velocity constants for Method A, referred to a perfect α-ray bulb of zero radius, no dead arm, and to the initial ordinates of the ionization curve for all three sets of α-particles are: Expt. 1*A*, $9.46 \times 1.0626 \times 1.0466 \times \frac{2.330}{2.659} \times 1.0488 = 9.67$; Expt. 2*A*,

[9] Lind, This Journal, **41**, 533 (1919).

[10] The reality of the recoil atom effect has been verified by new experiments which will be described subsequently. The new data are used for the corrections, but do not differ greatly from the previous results.

$$7.05 \times 1.0564 \times 1.0353 \times \frac{2.330}{2.645} \times 1.02647 = 6.97.$$

The velocity constants for pure α-ray effect corrected for volume of "magnetic capsule," and referred to initial ordinates on the ionization curve are: Expt. 1B, $6.65 \times 0.995 \times 0.916 \times \frac{2.33}{2.375} = 5.95$; Expt. 2$B$, $4.64 \times 0.996 \times 0.944 \times \frac{2.33}{2.398} = 4.24$.

The average path in Method A is the radius of the reaction sphere. Therefore, the average path in Method B is for Expt. 1 equal to $\frac{5.95}{9.67} \times r = 0.615\, r$, and for Expt. 2 equal to $\frac{4.24}{6.97} \times r = 0.609\, r$. These values are reported and discussed in the preceding paper.

It was found from the data obtained by methods described in Section 3, that 94.4% of the α rays get through the α-ray bulb. Assuming that the velocity of reaction is proportional merely to the linear path of the α ray in the gas phase, the velocity constants for Method A (no ionization considerations) are: Expt. 1A, $9.46 \times 1.030 \times 1.0488 \times 1.0626 \times 1.059 = 11.50$; Expt. 2$A$, $7.05 \times 1.024 \times 1.02647 \times 1.0564 \times 1.059 = 8.28$. The velocity constants for Method B are: Expt. 1B, $6.65 \times 0.995 \times 0.916 = 6.06$; Expt. 2$B$, $4.64 \times 0.996 \times 0.944 = 4.36$.

The average path in mixtures would be for Expt. 1 equal to $\frac{6.06}{11.50} r = 0.527\, r$, and for Expt. 2 equal to $\frac{4.36}{8.28} r = 0.527\, r$, or to the limits of accuracy, $0.53\, r$. This value has no reasonable interpretation and is discussed in the preceding paper.

Summary

For the corrections involved in Part III, a knowledge of the characteristics of the α-ray bulb as a radiator, as affected by the tip and neck, by the thickness of the wall and obliquity of passage of α particles through it, and by the diameter of the bulb (reducing it to zero dimensions in order to afford radiation from a point source) is necessary. In addition, the other corrections applying to the outer sphere itself are treated, such as the dead-arm correction and the change of ionization intensity with the pressure. The recoil atom effect is also used as a correction for the results, to reduce them to the same conditions as those obtaining outside the α-ray bulb, through which recoil atoms cannot penetrate.

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