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THE IONIZATION AND STOPPING POWER OF VARIOUS GASES FOR ALPHA PARTICLES FROM POLONIUM. II.

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Abstract

The ionizations of nitric oxide, nitrous oxide, and air were found to be strictly proportional along the range, while methyl iodide shows definite systematic deviations.

A method of determining *differential stopping powers* for α-particles has been applied to a number of gases. The results show that such unstable molecules as NO and $H_3C-N=NCH_3$ give strictly additive stopping powers. The stopping powers of all investigated gases are constant within one or two percent for the first 16 mm of range for α-particles from polonium. The method used by Gurney, and by Gibson and Gardiner, for determining stopping powers at the end of the range was investigated, and modified in some details. A series of stopping powers for each of four gases was obtained.

Straggling of α-particles in various gases. The portion of a Bragg ionization curve between the maximum and the last 1.5 mm is approximately a straight line. If the ionization curve for an individual α-particle is a straight line, and if the straggling of the α-particles is given by the equation $y=e^{(-z/\alpha)^2}/\pi^{1/2}\alpha$ (derived theoretically by Bohr and Flamm, and tested experimentally by Irene Curie, Briggs, and others), the end of the Bragg ionization curve is accurately reproduced. This has been shown by Henderson and by Irene Curie. The values of α which they used were obtained from other considerations. The inverse process, that of finding α from the end of the Bragg ionization curve, has been shown in this paper to give values of α for air which agree with those given by the other methods. This procedure is applicable to the study of straggling in any substance chemically non-reactive.

IT SEEMED desirable to investigate to what extent amounts of ionization and stopping powers of elements are changed in compound formation. Much work has been done and an additive law for atomic stopping power has been deduced by Bragg which holds in many cases within experimental error.[1] On the other hand the number of ions formed appears to be very largely conditioned by chemical composition. The hydrocarbons yield about 1.3 times as many ions as air, per α-particle, while H_2, CO, and CO_2 yield approximately the same number. Since the number of ions formed is the ratio of molecular ionization to stopping power, molecular ionization cannot be the sum of the atomic ionizations. It was hoped that a study of certain additional compounds might shed light on these points. With this purpose in view, we determined relative ionizations and stopping powers at the beginning and end of the range of α-particles from polonium in a modified Geiger apparatus similar to that used by Gibson and Gardiner.[2]

The only essential modification was the inversion of the perforated brass plates (*D*, Fig. 1 of preceding paper by Gibson and Gardner) between which

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[1] Rutherford, "Radioactive Substances and Their Radiations," Cambridge University Press, 1913.

[2] Gardiner, Ph. D. thesis, California 1926; Gibson and Gardiner, Phys. Rev. (preceding paper).

the mica sheet was clamped in order to diminish the depth of the part of the perforations above the mica sheet. As will be shown later, this change reduced to a negligible amount the error due to loss of ions in the perforations.

All the gases, with the exception of N_2O, were prepared by standard methods in vacuum-tight apparatus. Suitable drying agents were used and fractional distillations from ether-carbon dioxide mixtures and liquid air were employed where feasible. Condensable gases were never used without first ascertaining the absence of permanent gases. Reactive gases were kept condensed in liquid air. Nitrous oxide was taken from a commercial container, was passed through P_2O_5, KOH, and distilled after removal of any gases not condensable in liquid air. Azomethane was kindly supplied by Dr. Herman Ramsperger. Samples of the same gas prepared at different times always gave concordant results.

IONIZATIONS

Ionization curves for certain gases at low pressures in the upper chamber have been obtained and have been extrapolated to zero pressure. The resulting curves for air, nitrous oxide and nitric oxide show proportionality of their ordinates within experimental error (about one percent) all along the range. The curves given in Figure 1 show the ionizations given by these gases for equal pressures in the upper chamber.

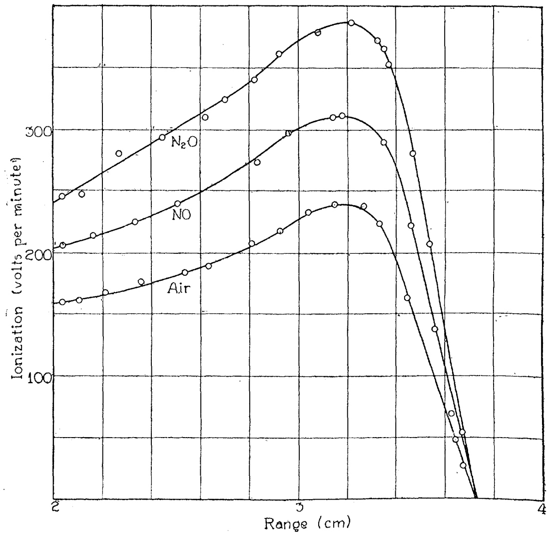

Fig. 1. Ionization in N_2O, NO and air for equal pressures in the upper chamber.

MOLECULAR AND TOTAL IONIZATIONS

Ionization curves of a series of gases at pressures of 2 cm in the upper chamber were also obtained. The depth of this chamber at this pressure of air is equivalent to slightly more than 2 mm at standard conditions. It is difficult to derive the curve for infinitesimal pressure in the upper chamber from such curves, but they furnish a means of getting the molecular ionization at a point far from the end of the range. The molecular ionization is the ratio of the ionization obtained from a number of molecules of gas to the ionization obtained from an equal number of air molecules. The molecular ionization divided by the differential stopping power gives the number of ions formed for unit decrease in range of the particle in air. That this quantity will be nearly the same for all points of the range and consequently equal to the "total ionization," follows from the rule stated in the preceding paper, that the energy loss per ion is constant along the range. The values for NO, N_2O, and CO_2 coincide exactly with the values given by Bragg for total ionization[3], as would be expected from the rule.

The stopping power in each case is one determined experimentally for a point about 6 mm nearer the source than the point at which the ionization was measured, but the stopping power is certainly constant to a greater distance from the source than this. Table I gives the experimental results.

TABLE I

Molecular ionization produced by α-particles in various gases.

Gas	Molecular Ionization	Molec. Ionization / Stopping Power
$(CN)_2$	1.855	1.000
HCN	1.205	1.126
CO	0.987	1.005
CO_2	1.520	0.996
N_2O	1.54	1.02
CH_3I	3.56	1.37
O_2	1.187	1.099
$CH_3N=NCH_3$	3.42	1.38
NO	1.29	1.29

The distance from the end of the range for which the above values apply is 1.68 cm.

STOPPING POWERS IN FIRST PART OF RANGE

The method employed was to use air at constant pressure in the upper chamber (2 cm in these measurements). Two ionization curves were then determined. In the first, air was used in the lower chamber; in the second, the gas whose stopping power was being determined. Corresponding points on the two curves were chosen such that the ordinate (ionization) of each was the same fraction of the maximum ordinate. The abscissas (ranges) were then plotted against each other. The slope of this curve is the differential stopping power of the gas. The thickness of the mica determined how near to the end of the range this method could be carried. The mica sheets varied from 1.5 to a little more than 2 cm air equivalent at standard conditions of temperature and pressure. The range of an α-particle from polonium was assumed to be 3.721 cm in these determinations.

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[3] Bragg, Studies in Radioactivity. p. 68, Macmillan 1912.

The calculated stopping powers in the table were obtained by taking $H_2=0.223$, $N_2^1=0.94$, $I_2^1=3.44$, $C=0.448$ (calculated from the values for O_2 and CO_2 in Table II.)

TABLE II

Stopping powers of various gases for α-particles.

Gas	Distance from source in cm	Stopping power Experimental	Calculated
H_2	1.20 to 1.64	0.223	
$CH_3N=NCH_3$	1.20 to 1.64	2.47	2.505
O_2	1.60 to 2.07	1.079	
O_2	1.80 to 2.21	1.076	
CH_3I	1.80 to 2.21	2.60	2.502
CH_3I	1.20 to 1.64	2.60	
N_2O	1.20 to 1.64	1.514	1.479
CO_2	1.20 to 1.64	1.527	1.527
CO	1.20 to 1.64	0.980	0.990
NO	1.20 to 1.64	1.006	1.009
HCN	1.20 to 1.64	1.070	1.030
$(CN)_2$	1.20 to 1.40	1.85	1.836

The stopping powers of the gases other than H_2, and $(CN)_2$ are probably correct for the ranges given, to within 1 percent. Cyanogen was prepared by dropping KCN solution on $CuSO_4$ and passing the gas repeatedly over P_2O_5, condensing it with liquid air, pumping off the non-condensable gases, and drawing the cyanogen used from an HCl-ice bath at $-20°C$. The determination seemed satisfactory at the time, but subsequent preparations of the gas by the same method failed to give reproducible results for stopping powers at the end of the range. This matter will be discussed again later.

The curves from which the above stopping powers were taken are straight lines within experimental error, so these values represent within one or two percent the stopping power for the first 1.6 cm of the range of α-particles from polonium.

Perhaps the most striking thing shown by this table is that NO and $H_3CN=NCH_3$ have stopping powers so near the values calculated on the basis of additivity of atomic stopping powers. The odd electron configuration manifestly has slight influence on the total amount of energy an α-particle loses when it is moving rapidly.

STOPPING POWER NEAR THE END OF THE RANGE

Four gases were investigated, hydrogen, methyl iodide, nitric oxide and cyanogen. The first two were run for comparison with the work of Gurney.[4] and of Gibson and Gardiner,[2] and the method, except for the modification already mentioned, was the same as theirs.

The air pressure in the lower chamber was fixed at some definite value, so that α-particles after traversing the air of the lower chamber and the mica sheet entered the upper chamber with a definite residual range. Varying pressures of air were introduced into the upper chamber. Each pressure yielded a certain rate of fall of the electroscope. Pressure and rate of forma-

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[4] Gurney, Proc. Roy. Soc. London **A107**, 340 (1925).

tion of ions were plotted against each other and a curve obtained which showed a flat maximum. This maximum corresponded to a gas pressure just sufficient to prevent the α-particles from striking the top of the box.

Table III

Ratio of maximum ionization in various gases to maximum ionization in air produced by α-particles at different points along their range.

Gas	Dist. from end of range in mm	Stopping power			$\frac{\text{Max. ionization gas}}{\text{Max. ionization air}}$
		Observed	Gurney	Gibson and Gardiner	
H_2	0.6			0.235	
	1.6			0.282	
	2.6			0.283	
	2.76	0.278			1.077
	3.5		0.309		
	3.6			0.293	
	3.82	0.303			1.076
	14		0.247		
	24.7 to 20.6	0.223			
	38 to 35		0.214		
CH_3I	0.6			2.10	
	1.6			2.29	
	2.14	2.56			1.42
	2.60			2.44	
	2.80	2.58			1.44
	3.60			2.61	
	3.68	2.64			1.43
	25.2 to 20.8	2.60			
NO	1.96	1.095			1.19
	2.68	1.047			1.237
	2.70	1.02			1.22
	3.18	1.035			1.21
	3.88	1.025			1.197
	4.42	0.96			1.23
	25.2 to 20.8	1.009			
$(CN)_2$*	1.94	1.739			1.02
	2.68	1.79			1.01
	2.68	1.57			1.03
	2.68	1.88			1.02
	3.15	1.84			1.05
	4.41	1.63			1.05
	2.52 to 2.08	1.85			1.03

* *The chronological order for the cyanogen determinations was* 1, 5, 2, 6, 3, 4.

Table III gives, in the first column, the gas, in the second the distance from the end of the range in millimeters, in the third, fourth and fifth columns the stopping powers obtained by the writers, Gurney, and Gibson and Gardiner, respectively. The sixth column gives the ratio of the number of ions formed in air to the number in the gas in question for the residual range for which the stopping power was determined. The checks with hydrogen are good, and this gas is the one which would be expected to be least affected by the loss of ions in the perforations in the original arrangement of the apparatus. The fifth column shows the stopping power of CH_3I falling off more rapidly than does the third. There is a real discrepancy here which probably is due to the loss of ions in the perforations in the earlier experiments. The results for NO seem to show a slight minimum in the stopping power curve.

The case of cyanogen is interesting in that it reveals a progressive increase in stopping power with time, the values 1.57, 1.79 and 1.88 having been obtained for the gas on successive trials for the same range. The explanation is probably polymerization. The gas was kept condensed in liquid air and allowed to vaporize from the solid when it was desired to admit it into the upper chamber. Every precaution was taken to insure drying and to remove any non-condensable gases, particularly when it was observed that the results were not reproducible. Cyanogen also appears to have a minimum stopping power near the end of the range.

Since the apparatus as used by Gibson and Gardiner[2] had, on the upper side, the thicker of the two plates which clamped the mica, the α-particles upon entering the upper chamber first pass through perforations a centimeter deep from which some of the ions formed failed to emerge. This caused an increasing number of the ions to be lost, probably by recombination as the pressure in the upper chamber was increased. The resulting curves showed a dropping off beyond the maximum which was calculable if the proper fraction of each hole was considered to contribute no ions. After inverting the plate, the resulting curves showed much less dropping off beyond the maximum, and a series of air curves made with different pressures of air in the lower chamber had maxima which were exactly superposable. This meant that the fraction of the total number of ions lost in the holes was negligible.

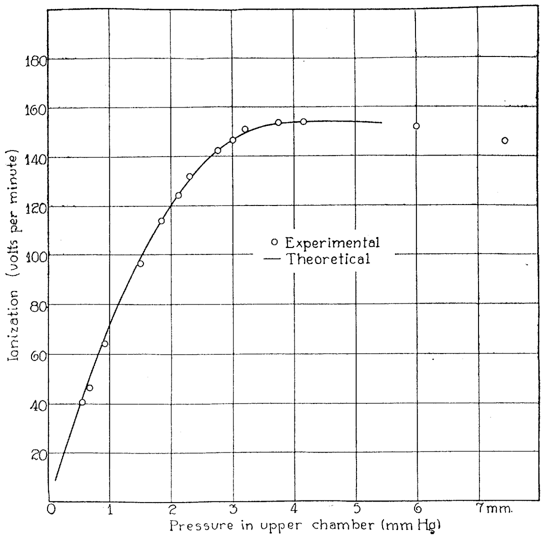

Fig. 2. Observed and calculated values of the ionization for various pressures in the upper chamber.

The truth of this was borne out strikingly by the precision with which the shape of the experimental curve was reproduced by integrating a theoretical ionization curve calculated from the Flamm–Bohr theory of straggling in a manner to be explained presently. The experimental points and the theoretical curve are shown in Figure 2.

The ionization curve obtained on the basis of the Bohr distribution of ranges was calculated as follows. The equation $y=e^{-(z/\alpha)^2}/\pi^{1/2}\alpha$ for the distribution of ranges about the mean was obtained theoretically by Bohr and experimentally verified by Irene Curie[5] and later by Briggs.[6]

In this equation y is the fraction of the total number of particles per mm of range having a range differing by z millimeters from the mean. Irene Curie determined $\alpha/37.21=0.0165$ for air, and as Briggs showed in the article quoted, this must be multiplied by $(1.18)^{1/2}$ for mica, giving $\alpha_1/37.21=0.0179$. In the stopping power experiments a second mica sheet was interposed immediately above the source. Thus slightly more than one-tenth of the range of an α-particle before reaching the upper chamber was through air, but this could not change α_1 essentially since nearly all the straggling occurs in the mica. Hence $\alpha_1=0.0179\times 37.21=2/3$ mm approximately, and the equation becomes

$$y=3e^{-9z^2/4}/2\pi^{1/2}\alpha.$$

But dJ/dR, the ionization per cm per sec. for a pencil of α-particles at some distance R from the most probable range, will be

$$dJ/dR=NS\int_{-R}^{\infty}3(R+z)e^{-9z^2/4}dz/2\pi^{1/2}$$

where the proportionality constant S multiplied by the distance $(R+z)$ of the individual α-particle from the end of its range gives the rate of ionization dJ'/dR for this particle. N is the number of α-particles in the pencil.

Integrating, this gives the expression

$$dJ/dR=(NS/2)\left[R(1+I_0{}^{3R/2})+2e^{9R^2/4}/3\pi^{1/2}\right]$$

where I is the probability integral. The quantity $(2/NS)(dJ/dR)$ was plotted and a curve obtained agreeing closely with the end of an experimental Bragg ionization curve. Integrating this curve we obtain the curve in Fig. 2. The constant $NS/2$ is determined by the condition that the integral to the end of the range from a definite abscissa must give the same total ionization as that obtained in the experimental curve.

CALCULATION OF α FROM THE IONIZATION CURVE AT THE END OF THE RANGE

The end of the range for a Bragg curve gives a means of calculating α in the equation

$$y=e^{-(z/\alpha)^2}/\pi^{1/2}\alpha\,. \tag{1}$$

This has been done for seven gases in the upper chamber, using curves obtained by Gibson and Gardiner. In these gases the α-particle had travelled through 1.71 cm of air (or slightly less) and then through about 2.0 equivalent cm of mica.

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[5] Irene Curie, Ann. d. Physique (10) **3,** 299 (1925).

[6] Briggs, Proc. Roy. Soc. London **A114,** 313 (1927).

The assumptions on which the deductions are based are that an individual α-particle has an ionization curve falling off linearly beyond the maximum; that straggling occurs according to equation (1) and finally that geometrical straggling, because of the form of the apparatus, may be neglected. This last point has been examined in detail. The second is in accord with both theory and experimental results. The first point is in keeping with the observed straightness of the ionization curve between the maximum and the point where an appreciable Bohr straggling would be expected to manifest itself.

From the above we have the relationship

$$\frac{dJ}{dR} = \frac{NS}{2}\left[R(1+I_0{}^{R/\alpha}) + \frac{\alpha}{\pi^{1/2}}e^{-(R/\alpha)2}\right]$$

and for $R=0$ we get

$$(dJ/dR)_{R=0} = NS\alpha/2\pi^{1/2}.$$

Also

$$\frac{d^2J}{dR^2} = \frac{d}{dR}\left[NS\int_{-R}^{\infty}(R+z)\,\frac{1}{\pi^{1/2}\alpha}e^{-(z/\alpha)^2}dz\right]$$

$$= NS\int_{-R}^{\infty}\frac{d}{dR}\,(R+z)\,\frac{1}{\pi^{1/2}\alpha}e^{-(z/\alpha)^2}dz + 0$$

For $R=0$ we have

$$\left(\frac{d^2J}{dR^2}\right)_{R=0} = NS\int_0^{\infty}\frac{1}{\pi^{1/2}\alpha}e^{-(z/\alpha)^2}dz = \frac{NS}{2}$$

Whence

$$\alpha = \pi^{1/2}\left(\frac{dJ/dR}{d^2J/dR^2}\right)_{R=0}$$

Thus α is simply the subtangent to the point on the curve corresponding to the end of the range, multiplied by $\pi^{1/2}$. The values obtained for the gases in the upper chamber plotted in Fig. 5 of the preceding paper are given in the following table. On the assumptions above stated they should all give the same value.

TABLE II

Values of the straggling constants for various gases.

Gas	Subtangent	α in mm
CH_3I	0.31	0.549
O_2	0.323	0.572
A	0.37	0.645
N_2	0.46	0.815
Ne	0.422	0.747
H_2	0.368	0.634
He	0.473	0.835
	Average	0.685

The average value 0.685 is to be compared with Curie's[5] value of 0.618 mm for air, and Brigg's[6] value of 0.666 mm for mica. It should be closer to the true value for air than for mica. The method is quite different from the cloud track method of Curie and the magnetic method of Briggs, and gives a rapid method for determining the straggling constant α with considerable accuracy for any gas placed in the lower chamber by simply obtaining accurately the ionization curve for the end of the range for some gas such as H_2 in the upper chamber.

Geometrical Straggling

The geometrical structure of the apparatus is the source of an inhomogeneity of ranges which we have called "geometrical straggling." This was the object of a detailed investigation, of which only the results will be given here. The geometrical straggling is small compared to the Bohr straggling and produces an average increase in the distance travelled by the α-particles of about 0.4 percent. The correction is too slight to interfere seriously with the determination of the straggling constant α from the end of the ionization curve, and affects the ranges in all gases by the same fractional amount so that it does not interfere with the conclusions drawn from the data obtained with this apparatus, in so far as relative values for the different gases are concerned.

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