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LII. The distribution of the active deposit of radium in an electric field.—II

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absorption experiments, and it is shown why an exponential law signifies heterogeneity of the rays. In the case of X rays scattering is in most cases so small compared with absorption that an exponential law probably signifies homogeneity.

In conclusion, the writer wishes to express his best thanks to Prof. Barnes for placing all facilities for this research at his disposal.

McGill University,
Montreal.

LII. *The Distribution of the Active Deposit of Radium in an Electric Field.*—II. By E. M. WELLISCH, Assistant Professor of Physics at Yale University*.

Introductory.

1. **T**HE experiments described in the present paper are a continuation of the investigation made by Wellisch and Bronson† on the distribution of the active deposit of radium in an electric field. In that investigation radium emanation mixed with air was introduced into a cylindrical condenser, and the relative amounts of active deposit which settled on the central electrode and on the case were determined after equilibrium had been established for different positive potentials applied to the outer electrode.

It was shown that the part of the active deposit which settled on the case (anode) was due to the diffusion of uncharged carriers; no evidence was found of the presence of negative carriers in any appreciable amount. It was found also that, when the applied potential was not too small, the distribution of the active deposit was independent of the quantity of emanation employed, and that the fraction of the total amount which settled on the cathode in general increased with increasing potentials, although under the most favourable conditions there was still about 10 per cent. which was deposited on the case. The passage of Röntgen rays through the gas during the exposure was found to be without effect on the distribution except when the applied potential was small, in which case the extra ionization produced by the rays caused increased recombination with the charged active deposit particles, and in this manner the cathode deposit was diminished. Finally, it was found that for potentials which were not too small the ratio of the

* Communicated by the Author.

† Wellisch and Bronson, *Phil. Mag.* [6] xxiii. p. 714 (1912).

equilibrium ionization currents in the gas for two different potentials was equal to the ratio of the corresponding cathode activities.

The main object of the present series of experiments was to extend the investigation in various directions; in particular, it was thought desirable to ascertain the effect on the distribution of employing a containing vessel of different dimensions and of mixing the emanation with gases other than air, and, in addition, to make a detailed investigation of the distribution when small potentials were employed. The main experimental results of the previous research have been confirmed, but the fresh results which have been obtained necessitate a modification of the theory which was suggested in explanation of the phenomena.

Experimental Procedure.

2. The method employed for ascertaining the distribution of the active deposit was the same as that which had previously been employed. The radium emanation obtained usually from a quantity of carnotite, or in some cases from an aqueous solution of a radium salt, was passed into the containing vessel and remained there under the desired conditions of potential, pressure, &c., until radioactive equilibrium was established; in general this period was about 3 hours. The emanation was blown out by means of a strong current of air from a force-pump, and the ebonite plug containing the central electrode was then removed. A fresh electrode was suspended in the vessel, and the ionization current due to the "case" activity was measured at 10 and 15 minutes after the emanation had been removed; the activity on the central electrode was measured by suspending it in a vessel of construction identical with that which contained the emanation, and observing the ionization current at 20 and 25 minutes after the emanation had been removed. The activity when in equilibrium with the emanation was then calculated in the usual manner.

The diagram of connexions is the same as that given in the previous paper, and is reproduced in fig. 1; in the present investigation R and R' were wire resistances of 50,000 and 10,000 ohms respectively. The Dolezalek electrometer had a platinum suspension, and with 120 volts on the needle the sensitiveness was 180 mm. per volt. C and B represent capacities which could be added to the system by means of the key K , and the total capacity of the system was then increased 21 times. A potentiometer device, not shown in the diagram, was employed when measurements of

the ionization current were made; this device enabled the range of swing of the electrometer-needle to be so adjusted that its mid-point coincided with the zero of the instrument,

Fig. 1.

a precaution which was especially necessary when the applied potential was small. Reference should be made to the previous paper for a more detailed account of the apparatus and method of procedure.

Experiments with a Cylindrical Condenser of Small Diameter.

3. In the previous experiments the greatest amount of active deposit that settled on the cathode was about 90 per cent. of the total; this occurred for a potential of 4000 volts. It was of interest to ascertain the effect of applying a large potential across a smaller distance so as to obtain very large values for the electric field. For this purpose two cylindrical vessels were constructed of the following dimensions:—

Height	140 mm.
Inner diameter	19 „
Length of central electrode	101 „

Radium emanation mixed with air at 1 atmosphere pressure was introduced into one of these vessels, and when a positive potential of 3000 volts was applied to the case it was found that about 86 per cent. of the deposit settled on the cathode. In all probability some part of the cathode activity made its way to the ebonite insulation, but there did not appear to be any gain in the cathode deposit as a result of decreasing the cross-section of the containing vessel.

Cathode Deposit in Dry Air at Different Pressures.

4. Throughout the remainder of the experimental investigation, use was made of the two cylindrical vessels which had been employed in the previous research. These vessels were identical in construction and the dimensions of each were as follows:—

Height (inside)	140 mm.
Inner diameter	58 „
Exposed length of central electrode	132 „
Diameter of central electrode . .	1.83 „ .

The inner electrodes were made longer than those which had previously been employed, and care was taken that no appreciable part of the active deposit was able to settle on the ebonite insulation. During the course of the present experiments a fact was noted which had previously escaped observation. Discrepancies, in general small, occurred in the values for the cathode deposit when the experimental conditions appeared to be identical. Repeated attempts to ascertain the cause of the discrepancies were for a long time unsuccessful, but finally it was ascertained that the inconsistent results arose from the presence of small quantities of water vapour in the gas. In the previous research a test had purposely been made to find the effect of neglecting to dry the gas with which the emanation was mixed; this test appeared to show that the cathode deposit was unaffected by omitting this precaution. However, the fallacy of this result was shown by more thorough investigation. The effect of water vapour is to diminish the cathode deposit, and is especially marked when the gas pressure is high and the applied potential fairly small; in these circumstances an amount of water vapour which was not sufficient to produce any perceptible increase in the recombination of the ions present in the gas might easily diminish the cathode deposit by 30 to 50 per cent. Further illustrations of this effect are given later, but in future experiments extreme care was

taken to dry the gas with which the emanation was mixed. This was done by passing the gas through several tubes containing P_2O_5 and glass-wool before it entered the testing vessel.

The following table gives the values obtained for the cathode deposit expressed as a percentage of the total deposit when the emanation was mixed with dried air at pressures of 210 mm. and 760 mm., and various positive potentials were applied to the case:—

Potential in volts.	Percentage cathode activity.	
	Air at 210 mm.	Air at 760 mm.
20.....	...	65.3
40.....	88.7	74.8
160.....	88.8	83.9
1030.....	89.2	89.2
2000.....	88.8	...
4000.....	...	89.2

These values have been corrected for the amount of uncharged deposit that diffuses to the cathode during the exposure; this correction was made by assuming that the uncharged deposit particles were distributed on the cathode and the case in proportion to the exposed areas, which were as 1:50. The figures given represent accordingly the number of positive carriers of activity expressed as a percentage of the total number of carriers.

In order to demonstrate experimentally that the activity which appeared on the anode was almost entirely due to the diffusion of uncharged carriers, several experimental determinations were made of the distribution of the active deposit when a large negative potential was applied to the case. As an example of the results obtained in this connexion, it was found that when the emanation was mixed with dry air at 1 atmosphere and with a negative potential of 160 volts, less than 2 per cent. of the total deposit appeared on the central electrode (anode), showing that no appreciable part of the active carriers are negatively charged.

For potentials greater than about 40 volts the percentage cathode activity is independent of the amount of emanation employed unless the amount be inordinately large; over the same range of potentials, moreover, it was verified that the ratio of the two ionization currents obtained for any two potentials was identical with the ratio of the corresponding

percentage cathode activities. The values obtained for the percentage cathode activity for air at 210 mm. pressure are greater than those obtained in the previous research. It is probable that this discrepancy is due to the fact that in the previous experiment some of the ebonite insulation was exposed to the emanation, so that the central electrode did not receive all the positive carriers. The insulation would probably act as a partial conductor, especially at the lower pressures when it would be exposed to the α radiation proceeding from a considerable distance. In the present experiment this source of error was carefully avoided, and the result appears to be that the same maximum value is obtained for the percentage cathode activity both for the lower and the higher pressure.

In the previous work the assumption was made that 100 per cent. was the limiting value which the cathode deposit approached as the potential was increased, and that even at the low pressures the saturation attained was merely apparent. This assumption was made chiefly as a result of the experimental observation that the percentage cathode activity was greater at the higher than the lower pressures. Since, however, it has now been shown that the percentage cathode activity has the same value (89.2) at the higher potentials for both pressures, it appears much better to regard this as the true limiting value. The gradual increase of the values for one atmosphere for potentials above 40 volts shows that the phenomenon of columnar recombination is present: the active deposit particle recoils into the gas after the expulsion of the α -particle from the atom of emanation and tends to recombine with the negative ions which it forms along its path.

Experiments made with a steel instead of a brass central electrode gave the same limiting value for the percentage cathode activity, indicating that this value does not depend upon the nature of the material of which the electrodes are composed.

At low pressures, as is well known, a considerable number of the active deposit particles may reach the walls of the containing vessel before their velocity is sufficiently reduced to enable them to be directed by the electric field. With air at a pressure of 6 mm. and with 180 volts the percentage cathode activity was found to be 66.7.

Experiments with Small Applied Potentials.

5. The experiments described in the preceding section refer to potentials for which the distribution of the active

deposit was independent of the amount of emanation employed. For smaller potentials the distribution depends considerably on the amount of emanation: this arises from the fact that with these potentials recombination can occur between the positive particles and negative ions which are produced in the volume of the gas, whereas for the larger potentials recombination can only occur to any appreciable extent with negative ions which are present in the same column as the active particle.

A number of experiments were performed to ascertain in what manner the cathode deposit depended upon the amount of emanation for any applied potential, and especially to see whether the distribution would vary in the same way as the ionization current which passed through the gas during the exposure.

In fig. 2 there are given two sets of curves which represent

Fig. 2.

the results obtained in this series of experiments. The abscissæ represent the ionization current in scale-divisions per sec. (with added capacity) due to the emanation and active deposit in equilibrium when a positive potential of 160 volts was applied to the case. Inasmuch as this potential afforded the same percentage (viz. 94.3) of the saturation current whatever amount of emanation was employed,

the abscissæ (denoted by I_{160}) serve as a measure of the saturation current.

The continuous curves in fig. 2 have as ordinates I_V/I_{160} , *i. e.* the value of the current obtained with V volts applied to the case expressed as a fraction of the current obtained when 160 volts were applied.

The broken curves refer to the active deposit and have as ordinates A_V/A_{160} , *i. e.* the cathode deposit obtained with V volts applied to the case expressed as a fraction of the cathode deposit obtained when a positive potential of 160 volts was applied. As mentioned above, the cathode deposit obtained for a potential of 160 volts was only 83·9 per cent. of the total amount, and the maximum amount obtainable on the cathode for very large potentials was 89·2 per cent. of the total.

By plotting the curves in this manner the two sets become comparable; the continuous curves afford a measure of the fraction of the total number of positive ions which reach the cathode corresponding to any potential V , while the broken curves similarly afford a measure of the fraction of the total number of positively charged particles which settle on the cathode.

A very large number of experimental results were used in order to plot the curves; for the sake of simplification the individual results are not recorded in the diagram.

The curves in fig. 2 all refer to the values obtained when the air with which the emanation was mixed was thoroughly dried, as described in Section 4. The effect of a small amount of water vapour was especially marked when the applied potential was small. In illustration of this point some of the results obtained for dried and undried air are recorded below:—

Air at 1 atmosphere : V = 8 volts	{		A_V	I_V/I_{160}	I_{160}
dried with special caution			38·4	·61	3·21
containing slight traces of water vapour	{		31·2	·61	3·21

It is worthy of notice that the presence of small quantities of water vapour does not appreciably diminish the fraction of positive ions which reach the cathode, whereas the effect on the number of positively charged deposit particles is considerable. It has for some time been known that water vapour is effective in causing increased recombination of ions, but the above results serve to show that the ions are not nearly so sensitive to the presence of vapour as the active deposit particles.

Referring again to the curves of fig. 2, it is seen that in general the "activity" curve for any given voltage lies below the ionization curve for the corresponding voltage. This is almost certainly to be ascribed to the increased recombination with negative ions which occurs with the active particles as compared with the positive ions, even when the air with which the emanation is mixed is thoroughly dried.

*Cathode Deposit for very Small Quantities of
Emanation in Dry Air.*

6. If the curves in fig. 2 are produced so as to intersect the axis of ordinates, we obtain points which afford a measure of the fraction of ions and of positively charged deposit particles which would be obtained by the application of the corresponding potential when the air in the ionization vessel contains only a very small amount of emanation. These points are plotted both for ionization current and activity as separate curves in fig. 3; they may be regarded as limiting

Fig. 3.

curves which correspond to the absence of volume recombination in the vessel even at the smallest potentials employed. The upward slope of the curves is due entirely to the fact that increasing potentials prevent more and more the recombination of the positive ions or particles with negative ions which are produced inside the α -particle column.

It will be seen from the curves that any given potential

brings over to the cathode a larger fraction of ions than of positively charged deposit particles ; at a potential of about 40 volts the two curves practically coincide. The difference between the two curves can be explained by supposing that the negative ions which are produced in the column recombine with the positively charged active particles with greater facility than with the positive ions.

*Emanation mixed with Carbon Dioxide, Hydrogen,
and Ethyl Ether.*

7. When the emanation was mixed with dry CO_2 at various pressures, it was found that the maximum value obtained for the percentage cathode activity was 80·7. In order to obtain this value with potentials less than 1000 volts, the pressure had to be less than about 150 mm.

When the emanation was mixed with dry hydrogen the maximum value obtained for the percentage cathode activity was 89·2, the same as that obtained with air. This value could readily be obtained with a potential of 160 volts and with hydrogen at a pressure of 1 atmosphere, showing that there is very little columnar recombination in this gas. Hydrogen was found to be particularly sensitive to the presence of small traces of water vapour; the effect of the water vapour was to increase the potential necessary to obtain the same limiting value.

Inasmuch as the presence of minute quantities of water vapour resulted in a marked diminution of the amount of active deposit which settled on the cathode, it became of interest to ascertain the percentage of positively charged carriers which would result from mixing the emanation with a vapour. For this purpose ethyl ether was chosen ; any gas which remained in the vessel was swept out by a stream of ether which had previously passed through P_2O_5 . The following results were obtained :—

Pressure.	Potential.	Percentage cathode activity.
mm.	volts	
82	160	6·4
85	1070	9·8
128	do.	10·0
235	do.	10·8

At the highest pressure there was a large current passing

through the vapour during the activation, due mainly to the fact that the ether being near the point of condensation was partly conducting; this conduction current may have been responsible for the increased amount of the cathode deposit at the highest pressure. Apart from this it appears that for ether vapour the limiting value of the cathode activity is approximately 10 per cent.

Summary and Discussion of Results.

8. When the emanation is mixed with any gas there appears to be a definite limit to the fraction of the active deposit which settles on the cathode. This limit is independent of the pressure of the gas provided it is high enough to prevent the deposit particles from recoiling on to the walls of the vessel; it is in general dependent on the nature of the gas. This limiting value is in general obtained only with large potentials; with smaller potentials the fraction of the cathode deposit is decreased as a result of columnar recombination of the positively charged particles with negative ions; and with very small potentials the charged particles recombine with negative ions in the volume of the gas. Small traces of water vapour have a considerable effect in diminishing the number of positively charged particles: the water vapour appears to be effective in bringing about increased recombination both volume and columnar between the charged particles and the negative ions.

It has been shown in Section 5 that even in air which has been thoroughly dried the recombination between the charged deposit particles and the negative ions is greater than the recombination between the positive and negative ions. This result, which is in all probability to be ascribed to the larger size and mass of the deposit particles, is not in accord with the experimental result of H. W. Schmidt*, who came to the conclusion that as far as recombination and mobility are concerned the active particles behave as positive ions.

The process which accompanies the deposit of the active particles on the cathode appears to be most suitably explained in the following manner. At the moment of expulsion of the α -particle from the atom of emanation the residual part recoils into the gas; in air at a pressure of 1 atmosphere the range of this recoil atom has been shown to be about $\frac{1}{10}$ mm. As it moves through the gas the recoil atom produces a large number of ions, and in the act of ionization it is possible that the recoil atom may lose its positive charge. On the other hand, recoil atoms which at any time are uncharged

* H. W. Schmidt, *Phys. Zeit.* ix. p. 184 (1908).

may regain a positive charge, so that if we consider a large number of recoil atoms there will at any given moment be a certain fraction which carry a positive charge, the remainder being practically all neutral. The process is in many respects similar to that which is known to occur in the case of canal rays. During the motion of recoil the atom is practically unaffected by any applied electric field, so that initially the relative number of uncharged and charged recoil atoms is independent of the applied potential. However, when the recoil atom has reached the end of its path, if it be positively charged it may lose its charge by recombination with a negative ion formed in the column; this recombination can be prevented by increasing sufficiently the applied potential. Moreover, for small applied potentials a positively charged recoil atom may recombine with a negative ion in the volume of the gas.

When both columnar and volume recombination are avoided by the application of a sufficiently high potential, the distribution of the active deposit on the electrodes is determined entirely by the relative number of charged and uncharged carriers resulting from the recoil of the atoms of RaA in the gas. Under these circumstances we should expect that the distribution would be independent of the pressure of the gas because the recoil atom will meet the same number of gas molecules before it is brought to relative rest. Of course, if the pressure is too low an appreciable number of active deposit particles will recoil on to the walls of the vessel, and in this manner the cathode deposit will be diminished.

Although nothing has been established in this research with regard to the velocity of the recoil atoms when moving under the influence of an electric field, nevertheless there is distinct evidence that, so far as diffusion and recombination are concerned, the recoil atoms behave differently from the positive gas ions. It has been shown in Section 5 that, when recombination occurs between negative ions on the one hand and positive ions or positive recoil atoms on the other hand, a considerably smaller fraction of recoil atoms than of positive ions is received by the negative electrode. This is especially the case with moist gases, but even in gases which had been dried with the utmost care the difference is well marked. An examination of the curves of fig. 3 seems to afford further information in this connexion. The curves may be regarded as giving the fraction either of positive ions or of positively charged recoil atoms that is received at the negative electrode for any given potential, volume recombination being supposed to be entirely absent. It will be

noticed that these curves cut the axis of ordinates at the points marked .7 and .4, these points representing respectively 66 per cent. of the total number of positive ions and 38 per cent. of the total number of positively charged deposit particles. This type of curve has already been treated by Wellisch and Woodrow* for the case of the columnar recombination resulting from α -particle ionization. It was shown by them that the ordinate of the point of intersection represents the fraction of the total number of ions which escapes from the α -particle column as a result of molecular agitation and diffusion. Inasmuch as volume recombination is absent, these ions are brought over to the electrodes by a very small electric field. If we draw through the point of intersection a straight line parallel to the axis of potential, and if we refer the curve to this straight line as a new axis of potential, then the new ordinates will indicate to what extent the electric potential is effective in preventing recombination between those ions which still remain in the column after the initial diffusion has occurred. If we treat the curves of fig. 3 in a similar manner we see that, whereas in the vessel employed 66 per cent. of the positive ions on the average escaped from the α -particle column, the corresponding figure for the positively charged recoil atoms was only 38 per cent. This slowness with which the recoil atoms diffuse is readily ascribable to their relatively large size and mass. Of those ions and recoil atoms which do not escape by diffusion from the column, approximately the same fraction is brought over by any given potential; it seems that there is here some compensating influence at work, probably the greater tendency of the recoil atoms to recombine with negative ions is partly balanced by the smaller number of encounters with these ions.

The existence of a definite limiting value to the percentage cathode activity has been ascribed above to a continual process of gain and loss of charge which occurs during the recoil motion of the active deposit particle. It is to be expected that this limiting value will depend upon the nature of the gas into which the particle recoils; the experimental determination showed that this was in general the case, although the limiting value for hydrogen was within the limits of error the same as that for air. The fact that the value for ether is as small as 10 per cent. is surprising, and is in all probability to be ascribed to the ease with which the molecules of ether are ionized.

* Wellisch and Woodrow, *Phil. Mag.* Sept. 1913.