The Distribution of the Active Deposit of Radium in an Electric Field.—By G. H. Henderson, M. A., Instructor in Physics, Dalhousie University,* Halifax.

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I. Introductory.

The experiments described in this paper were suggested by the results of a previous investigation, by the writer[1], of the analogous problem for thorium. In the former paper it was shown that all the active deposit particles (or rest-atoms) of thorium could be collected on the negative electrode in a strong electric field. Hence all the rest-atoms, at least at the end of their recoil, are positively charged.

In the case of radium, however, early investigators found that only from 90 to 95% of the total active deposit was positively charged. But their results were little more than qualitative in nature. More recently Eckmann[2] came to the conclusion that 98% of the rest-atoms were positively charged and the remaining 2% negatively charged. The experiments of Walmsley[3] seemed to show that, even in the strongest fields, the fraction of the rest-atoms which was positively charged reached a maximum, leaving a small fraction uncharged. The methods of both experimenters, however, are open to very serious objections; the most important is the fact that they both used a cylindrical testing vessel with a central electrode, and then assumed that the total active deposit was the sum of the deposits obtained on it when first cathode and then anode. They thus neglected the considerable amount of deposit which collected on the case of the vessel itself.

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*Contributions from the Science Laboratories of Dalhousie University [Physics].

1. Henderson, Trans. N. S. Inst. Sci., XIV. pt. 1, p. 1.
2. Eckman, Jahr. der. Radioakt., May, 1912.
3. Walmsley, Phil. Mag., Oct., 1914.

The only apparently satisfactory results which remained were those of Wellisch[4]. He found that there was a limiting value to the fraction of the rest-atoms which could be concentrated on the cathode, as the strength of the field was increased. Later[5], he gave .882 as the value (corrected for diffusion) of this limiting fraction when the radium emanation was in an atmosphere of dry air. He found similar limiting values of .882 and .789 when the emanation was in atmospheres of hydrogen and carbon dioxide respectively. He also found that all the rest-atoms were uncharged in an atmosphere of ether vapor, as was found by the writer in the case of thorium. Several experimenters, Wellisch and Bronson[6], Wellisch[4], and Walmsley[3], have shown quite conclusively that no appreciable fraction of the rest-atoms is negatively charged. Hence, according to Wellisch's results, 11.8% of the rest-atoms are formed uncharged in dry air.

In the writer's experiments on thorium it was found that the charged condition of *al* the rest-atoms was the *same* in a pure gas (*i. e.* either all positive or all neutral), and that in a mixed atmosphere the fraction of the rest-atoms positively charged depended on the relative proportions of the constituents of the mixture. The explanation of this seemed to be that the charged condition of the rest-atom depended on what kind of molecules surrounded it.

No such simple explanation seemed applicable to the results of Wellisch. It, therefore, seemed desirable to investigate further the behavior of the rest-atoms of radium.

It was suggested by the writer in the previous paper that the fact that there seemed to be a limit to the fraction of the rest-atoms one could collect on the cathode was due to the fact that air was a mixture. This formed the starting-point of the present investigation. It might perhaps be

3. Walmsley, Phil. Mag., Oct., 1914.
4. Wellisch, Am. Journ Sci., Oct. 1913.
5. Wellisch, Am. Journ. Sci., Oct. 1914.
6. Wellisch and Bronson, Am. Journ. Sci., May, 1912.

expected that in an atmosphere of pure nitrogen, say, all the rest-atoms would be similarly charged. Experiments carried out in atmospheres of pure nitrogen and oxygen gave approximately the same fractions positively charged as for air, which disposed of this suggestion.

A possible explanation of the results obtained by Wellisch might be found in the type of vessel used. In these investigations Wellisch, as well as the others, used as a testing vessel a cylindrical case with an insulated rod fixed centrally inside it, case and rod serving as electrodes. The great disadvantage of this type of vessel is its non-uniform field. A simple calculation will show that most of the drop in potential between the electrodes takes place near the rod. The presence of ionization still further weakens the potential-gradient in the body of the vessel and increases it near the rod. In the corners at the ends of the cylinder and near the insulating plug, the field is much weaker than even in the body of the vessel. Now, although the average value of the field within the vessel may be such as apparently to prevent recombination, there is a part of the volume of the vessel in which the field is very much weaker than the average and in which recombination might take place to a considerable extent.

To avoid this and other difficulties, the vessel used in the present experiments consisted essentially of two parallel plates surrounded by guard-rings. This type of vessel gives a much more uniform potential-gradient than the cylindrical type. The guard-rings prevent distortion of the field at the edges of the electrodes and also allow the use of insulating material on their outer edges without effect on the field between the real electrodes.

II. Apparatus and Method.

The testing-vessel used in these experiments was a slight modification of the "second type" of vessel, described

previously by the writer. It is shown in plan and elevation in Fig. 1.

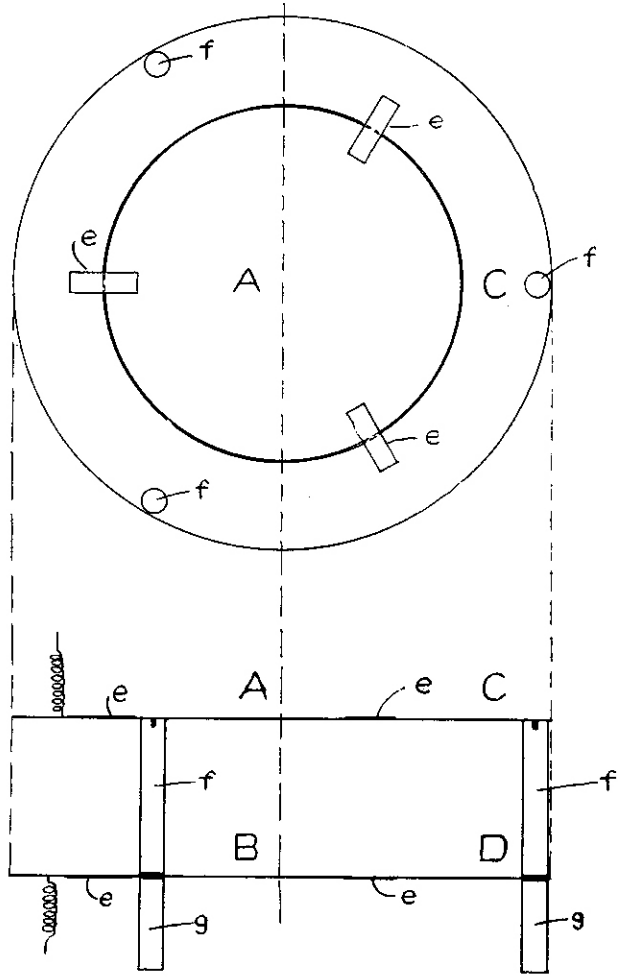

FIG. 1.

The electrodes A and B were two circular brass plates 7.00 cm. in diameter, supported on guard-rings C and D by small lugs e fixed to A and D. The guard-rings, also of

brass, were 7.10 cm. in internal and 10.5 cm. in external diameter. The guard-rings C and D were held parallel 3.00 cm. from one another by three glass rods f. The glass rods G supported the whole on the plate of an air-pump, to which the lower plate was electrically connected. The glass rods F and the outside edges of the guard rings were coated with paraffin to prevent electrical leakage, but no insulating material was near the field between A and B.

To prevent diffusion of recombined rest-atoms from above or below the plates, cotton wadding was placed over C and under D to close up the small space between the rim of the guard-ring and the bell-jar. This was the only essential modification of the previous vessel. The effect of the wadding will be referred to later.

In filling the bell-jar with the required gas the former was exhausted to at least 1 or 2 mm. pressure and the gas was then allowed to pass in through a tube of $CaCl_2$ and a tube of P_2O_5. The bell-jar was again exhausted and the gas allowed to enter a second time. The radium preparation and two small dishes containing P_2O_5 were always kept in the bell-jar, being placed above A.

In this condition the apparatus was allowed to stand from 12 to 24 hours in order to allow sufficient emanation to collect. It should be noted that the greatest quantity of emanation present at any time was minutely small, very much less than that employed by Wellisch. This small quantity would not cause the potential-gradient near the plates to be much greater than that in the body of the vessel. Hence the potential gradient would not be very ununiform. For the last 4½ or 5 hours' exposure of the plates to the emanation the desired potential difference was applied between them. Above a potential difference of 450 volts, i. e. above a potential gradient of 150 volts per cm., the potentials were supplied by a Wimshurst machine run by an electric motor. The values of the potentials obtained

in this way were calculated from the length of the spark gap, by use of the formula:

$$V = 1500 + 30000d$$

where V is the potential difference in volts and d is the length of the spark gap in cms. When the bell-jar was removed the emanation was allowed to escape before the potential was withdrawn. The activities of the plates were then measured in an α-ray electroscope, at from 10 to 25 minutes after the escape of the emanation. The procedure was much the same as that described for thorium in the previous paper, except that correction was, of course, made for decay during the interval of time between removal of the emanation and measurement of the activity.

III. Experimental Results.

While carrying out experiments in air and other gases it was found that a much higher potential-gradient was required to bring over to the cathode a certain percentage of radium rest-atoms, than was needed to bring over the same percentage of thorium rest-atoms. A series of experiments was then made in dry air using high potential-gradients. The results obtained are given in Table I. The first column gives the potential-gradient in volts per cm. The second gives the percentage cathode activity, i. e. the percentage of the total activity collected by the cathode. The percentage of the total positively charged is obtained by subtracting from 100 twice the percentage found on the anode, as there is also deposited on the cathode by diffusion an amount of activity equal to that deposited on the anode. The error in the percentage cathode activity, which depends largely on the error made in measuring the anode activity, is probably somewhat less than 1%. The values of the percentage cathode activity given in Table I are each the mean of several values calculated from different observations.

TABLE I.

Volts per cm.	Percentage cathode activity.
13	92.7
25	93.5
80	94.8
150	94.9
1000	96.2
2000	96.4
4000	96.9
12000	97.8

These results differ in two respects from those found by Wellisch. In the first place a considerably larger fraction of the total activity is found positively charged at the higher voltages than the results of Wellisch indicated. Secondly, there appears to be no limiting value to the percentage brought to the cathode. As the potential-gradient is increased this percentage gradually, though slowly, increases. Thus, the evidence of Wellisch to the contrary, one seems justified in concluding that all the rest-atoms could be brought to the cathode in a sufficiently strong field, and hence that all the rest-atoms are initially positively charged. It is further to be noticed how much more difficult it is to bring over to the cathode the rest-atoms of radium than those of thorium under similar conditions. To obtain 97.8% of the thorium rest-atoms on the cathode a potential-gradient of only about 30 volts per cm. would be required, as compared with a potential gradient of 12,000 volts per cm. in the case of radium. The ionization between the plates was of the same order of magnitude in the two cases.

That the value previously found for the percentage cathode activity of radium was but little above that obtained

by Wellisch, must be ascribed to the fact that in the earlier types of vessels there was no means of preventing the recombined rest-atoms from diffusing into the vessel proper and settling on the plates. That such an effect was appreciable, even in the present vessel where there was little space between the edges of the guard-rings and the bell-jar, was shown in the following manner. Two tinfoil screens were arranged to slip over the plates while in the electroscope. One exposed only the outer part of the anode plate, while the other exposed an equal area at the centre of the anode. If diffusion of recombined rest-atoms took place from outside one would expect to find more activity per unit area near the edge than near the centre of the plates. This would of course be noticeable on the positive plate only. Experiments were performed, leaving out the cotton wadding previously referred to. It was found that the activity near the edge of the anode plate was about 1.4 times that on an equal area near the centre. Similar experiments carried out with the wadding in place gave the activity near the edge practically the same as that on an equal area near the centre.

A few experiments were carried out in atmospheres of hydrogen, carbon dioxide and sulphur dioxide, with the results shown in Table II.

TABLE II.

Potential gradient	Percentage Cathode Activity.		
Volts per cm.	H_2.	CO_2.	SO_2.
150	95.3	89.9	....
4000	96.2	93.1	93.2
12000	....	93.9	....

In hydrogen at a potential gradient of 150 volts per cm. more of the rest-atoms can be collected on the cathode than in air while at 4000 volts per cm. a smaller percentage is collected by the cathode than in air with the same field. With this intense electrical field it is difficult completely to eliminate brush discharge. This would produce water vapor by causing the hydrogen to combine with the traces of oxygen present. That some moisture was present seemed evident from the fact that the P_2O_5 in the vessel was affected to a much greater extent than under a low potential-gradient, or n any other gas tried, though the same precautions were taken in all cases to dry the gas before admitting it to the vessel. Very small quantities of moisture are known to lower considerably the percentage cathode activity. This would seem to explain the low values of this percentage found in hydrogen with high potential gradients. If the effect of moisture could be eliminated it seems probable that a higher percentage cathode activity would be obtained in hydrogen than in air at all potential gradients. In hydrogen as in air the conclusion seems justified that at a sufficiently high potential-gradient all the rest-atoms could be collected on the cathode.

In carbon dioxide and in sulphur dioxide, it is much more difficult to bring over the rest-atoms to the cathode than in air. It will be noticed that both carbon dioxide and sulphur dioxide at room temperatures are on the border line between vapors and gases. To test if any abrupt change in the behavior of the rest-atoms takes place near the critical temperature, experiments were carried out in carbon dioxide at temperatures of 4°C., 19°C., and 36°C. It seemed a little more difficult to bring the rest-atoms over to the cathode at the lowest temperature, but the difference was very slight.

The fact that the percentage cathode activity is increasing so slowly when it is as low as 93% should perhaps make

one hesitate to conclude that all the rest-atoms could be collected on the cathode with a sufficiently high potential-gradient However, it seems unnatural to suggest any limit other than 100 for the percentage cathode activity, especially in view of the fact that with the highest voltages the percentage was still increasing. Therefore, in the absence of contradictory evidence, it seems reasonable to conclude that in sulphur dioxide and in carbon dioxide all the rest-atoms of radium are initially positively charged.

IV. Conclusions.

In atmospheres of air, hydrogen, carbon dioxide, etc., all the rest-atoms of radium appear to be initially positively charged.

So far no gas or vapor has been discovered in which the radium rest-atoms are not all similarly charged. In this respect the behavior of the rest-atoms of radium is the same as that of the rest-atoms of thorium. The slight differences in this respect which do exist are differences of degree and not of kind.

In a future paper the writer hopes to give some discussion and explanation of these experimental results.

I wish to express my gratitude to Dr. H. L. Bronson for his continual inspiration and advice throughout this investigation.