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THE DISTRIBUTION OF THE ACTIVE DEPOSIT OF THORIUM IN AN ELECTRIC FIELD.

—BY G. H. HENDERSON, B.A., B. Sc., *Instructor in Physics*, Dalhousie University,* Halifax.

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The problem of the distribution of the active deposits of radio-active substances in electric fields is an old one and was early investigated by Rutherford[1], Russ[2], Kennedy[3], and others. In much of this early work no attempt at precision was made and a considerable part of it was rendered of doubtful value by faulty experimental arrangements. The problem in the case of radium was more recently taken up by Wellisch and ronson[4] and by Wellisch[5] and in the case of actinium by Walmsley.[6]

It was felt that a similar investigation of the active deposit of thorium was timely and might throw further light on the mechanism producing the charged condition of the active deposit particles and on the conditions which affect their distribution in electric fields.

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

1. Rutherford, Phil. Mag., Feb. 1900, Jan. 1903.
2. Russ, Phil. Mag, June , 1908.
3. Kennedy, Phil. Mag., Nov. 1909.
4. Wellisch and Bronson, Am. Journ. Sci. XXXIII, May 1912.
5. Wellisch, Am. Journ. Sci. XXXVI., Oct. 1913.
6. Walmsley, Phil. Mag., Sept. 1913.

Practically all the testing vessels used in the above researches were cylindrical cases having an insulated rod in the centre. These vessels have the disadvantages of having an ununiform field and of presenting surfaces of unequal area to the active deposits. Therefore it was decided to try vessels having parallel plate electrodes. It is important that the testing vessel be so constructed that the active deposit particles or "rest atoms" would be deposited only on the electrodes and none on other parts of the vessel where they would remain unmeasured. To accomplish this several types of vessels were used, only two of which need be described. The construction of the first type is shown in Figs. 1 and 2. The electrodes were cut out of zinc as shown in Fig. 1. The flaps

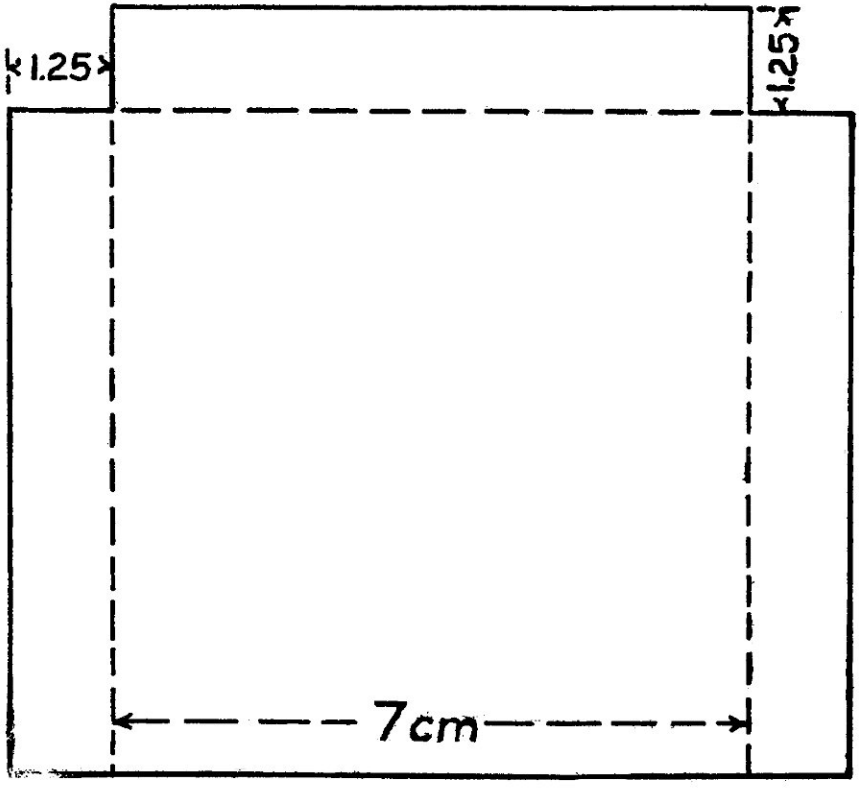


FIG. 1.

were folded over on the dotted lines forming a shallow box one end of which was missing. Two of these electrodes, their hollow sides facing one another, were slipped open ends downwards into a wooden case. The electrodes and case set up ready for an exposure are shown in cross section in Fig. 2. A A are the electrodes, B B is the case, C C are wooden pegs fixed in the sides of the case to separate the electrodes The edges of these were .3 cm. apart making the main parts of the plates 2.8 cm. apart. D is the bottom on which the case rests. It projects up a short distance inside B. The top of this projection is slightly hollowed out, forming a shallow trough G on which lay the thorium hydroxide, which was used as source of the thorium emanation. The plates rest on the rim of the trough. E E are leads and F is the cover of the box. All the wooden parts of the apparatus were boiled in paraffin to secure good insulation. Owing to the long period of thorium B (10.6 hours) the experiments necessarily proceeded with some slowness. Exposures varying from 6 to 36 hours were made, about 24 hours being usual. The plates were then removed from the vessel and their activities measured. As the decay curve for thorium active deposit of long exposure is practically flat for the first half hour, no correction for decay during the time of measurement was needed, and even with exposures as short as 6 hours only a small correction was required. This has been applied when necessary. The activity of the plates was measured by an α ray electroscope, similar to that described by Rutherford.[1] The earthed case of the electroscope was surrounded by a second case of cardboard with glass windows, to protect the instrument from extraneous effects of air currents and temperature. A water filter was als used to eliminate the heating effect of the lamp. The gold leaf was charged to 200 volts and its rate of fall was observed through a tele-microscope. The time required to pass over a certain

1. Rutherford, Radio-Active Substances, p. 90, fig. 12.

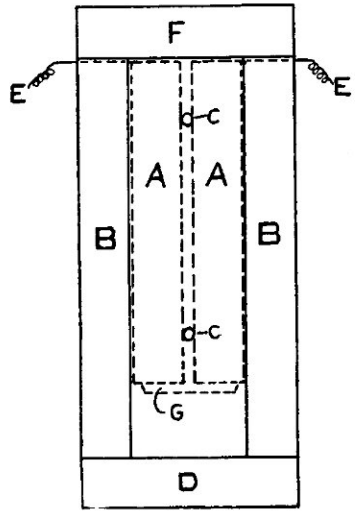


FIG. 2.

number of scale divisions was measured with a stop watch. This range of scale was kept the same for any given experiment. A correction for the natural leak of the instrument was always applied. This correction was usually a small fraction of the anode activity.

A typical set of measurements is given below:

Box 8 Plates 10 Voltage = 40.4. Dry air over H_2SO_4. Set up 2:15 P.M. Oct. 25, 1914.

Plates removed 9: 01 A.M., Oct. 26, 1914.

Plates.	Period.	Divisions.	Time.	Rate.	Cor. Rate.
10–	25.6″	60–50	9: 05 A.M.	.3906	.3880
10+	9′ 55.0″	60–50	9: 16 A.M.	.0168	.0142

Percentage cathode activity = 96.5.

The first line gives the conditions under which the exposure took place. The first column shows the electrode measured. The column marked "Period" gives the time taken by the gold leaf in moving over the scale divisions indicated in the next column. The column marked "Time" shows the time of conclusion of each measurement. The next column shows the rate of fall of the needle, while the last column shows this rate corrected for natural leak, and the decay when necessary. The corrected rate is a measure of the activity of the plate. The percentage cathode activity

$$= \frac{\text{Cathode activity} \times 100.}{\text{Cathode activity} + \text{anode activity.}}$$

The error in the values of the percentage cathode activity was mainly that made in measuring the anode activity. This error was seldom as much as 2 per cent. The readings were usually repeated several times and the mean taken.

An objection to these vessels might be raised, namely, that the plates are very close together at the edges, and that this distorted field was the determining part of the field. That this was not the case was shown as follows:

An exposure was taken as usual. The activities of the plates were measured as usual. Then the plates were covered with a zinc screen neatly covering the whole of the plates except a part in the flat portion of the plate where the screen was cut out, and which could expose a portion of either the top or bottom half of the plate. The size of this opening was 5.8 x 2.8 cm.

The activities of the plates were then measured with the lower and upper portions exposed. he results follow: Box 6. Plates 15. Volts 2.1. Laboratory air.

Whole Plates.

Activity on cathode = .604.*
" " anode = .294.
Percentage cathode activity = 67.3
Total activity on two plates = .89

Lower Portion Exposed.

Activity on cathode = .228.
" " anode = .114.
Percentage cathode activity = 66.7
Total activity = .33.

Upper Portion Exposed.

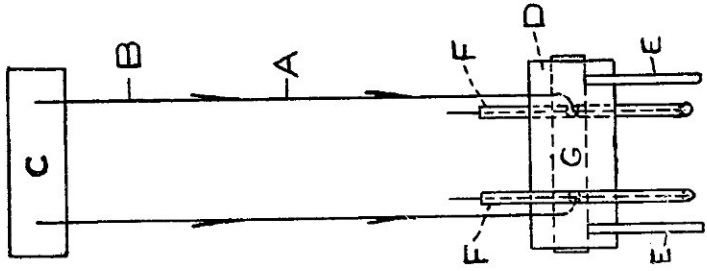
Activity on cathode = .157.
" " anode = .076.
Percentage cathode activity = 67.3.
Total activity = .23.

The size of the opening was about 22% of the total area of the plates including the sides. The relative total activities would show that the density of active deposit on the edges was less than that on the flat portion of the plates.

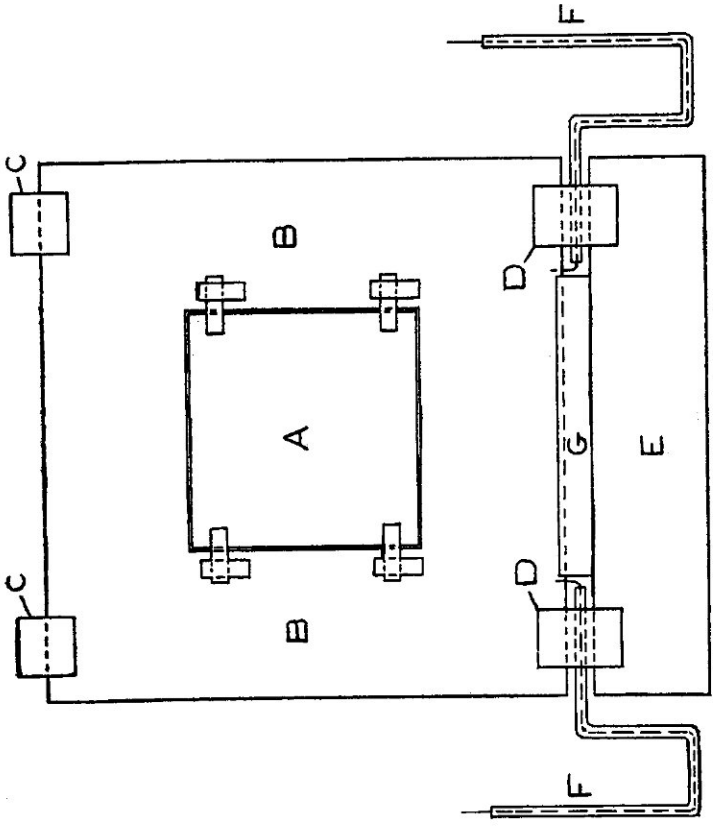
In general, fairly satisfactory results were obtained with this type of vessel. However, some discordant results were obtained which were hard to explain, but it was thought that the large amount of insulating material surrounding the electrodes might be the cause of these irregularities.

A second type of vessel was therefore constructed which avoided this difficulty as far as possible. End and side views of this vessel are shown in Fig. 3. The plates, which were of zinc, were made on the guard ring principle. The main plates, A, 5.8 cm. square, were slipped into place and held there by short lugs which engaged with clips on the back of the guard plate, B. The guard plates were 13 cm. square with an opening of 5.9 cm. square in the centre to receive the

*Arbitrary units.



End View—Fig. 3.



Side View—Fig. 3.

Fig. 3.

plates, A. The guard plates were kept 3.0 cm. apart by wooden blocks C and D at top, and bottom. The latter were supported on glass legs, E. The apparatus was set in a shallow glass dish and was covered by a rectangular glass jar. Control of the atmospheric conditions of the exposure was obtained by sealing the bell jar with a layer of liquid in the shallow glass dish. To secure dry air, concentrated sulphuric acid was the liquid used. In experiments on the effect of water vapor, water replaced the acid. For other gases a mercury seal was used to keep the desired vapor in the vessel. The thorium hydroxide was placed in a shallow wooden box G, under the plates and supported by the glass legs E. Connections with the plates were made by wires passing through glass tubes, F F, bent to pass under the bell jar through the liquid.

A number of advantages can be claimed for this type of vessel in studying the distribution of the active deposits.

The field is practically uniform over the plates A, any distortion of the field at the edges being eliminated by the guard plate.

No insulating material is near the electrodes.

Equal areas of electrodes are presented to the active deposit.

The activities of the two plates are measured under identically the same conditions, in the most simple manner, by a gold leaf electroscope.

Good control of gaseous conditions can be secured.

A series of observations was made with applied potentials varying from 6 to 12,000 volts. The highest voltage was obtained by the use of a Wimshurst machine run by an electric motor. The length of the spark gap was .35 cm. and the diameters of the knobs 3.0 cm. and 1.5 cm., giving an estimated voltage of 12,000. The air was dried by standing over concentrated sulphuric acid. The results obtained are shown in Table 1 and plotted in curve A, Fig. 4.

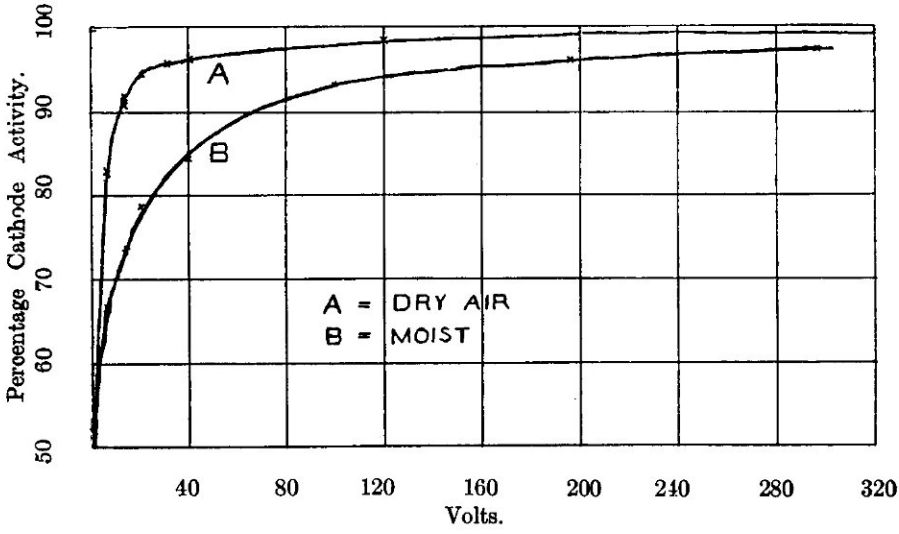


FIG. 4.

TABLE 1.

Volts.	Percentage cathode activity.
6.4	82.9
14.1	91.4
14.0	92.0
14.1	91.0
20.4	94.7
32.0	95.9
40.4	96.5
120.0	98.6
12,000.0	99.8

It will be seen that this curve has the general shape of the ionization saturation curve. The percentage cathode activity at 12,000 volts is the mean of three observations, in one of which the activity of the anode measured less than the natural leak. As this value differs from 100 by less than the observational error, there is no evidence to show that initially any fraction of the rest atoms is neutral. It must be understood that the use of the word initially, as in this instance,

refers only to a time after that infinitesimal fraction of a second during which recoil is operative. This curve differs in two respects from that found by Wellisch and Bronson for radium. (1) It reaches approximate saturation at much lower voltages. (2) There seems to be no evidence that any fraction of the rest atoms is initially unchanged. Experiments showed that the dimensions and form of the testing vessel and the concentration of the rest atoms are largely responsible for (1), and that approximate saturation in air is reached with about the same potential gradient for thorium as for radium.

Thus the evidence in the case of dry air would indicate that initially all the rest atoms of thorium are positively charged and that all the activity on the anode at low voltages is due to these rest atoms losing their charge by recombination with negative ions in the air.

It was found by Wellisch and Bronson that 10.4 per cent. of radium rest atoms were initially uncharged in dry air. Experiments were made to test whether this difference between radium and thorium was due to the type of vessel used.

The percentage cathode activity in the case of radium was measured with parallel plate vessels. The source of radium emanation used was a layer of radium chloride deposited on a sheet of aluminum. Preliminary measurements with the first type of vessel described, showed that this fraction was of the same order as that found by Wellisch and Bronson for cylindrical vessels. Later the experiment was repeated using the second type of vessel and a potential of about 12,000 volts supplied by the Wimshurst machine. The percentage cathode activity was found to be 5.2. This would make the total fraction unaffected by the electric field 10.4 per cent. in good agreement with the latest value (11.8) found by Wellisch[1].

1. Wellisch, Phil. Mag. Oct. 1914.

The distribution of the thorium active deposit was also measured in cylindrical vessels. The vessels were of the usual type. Air dried by passing over phosphorus pentoxide was passed over thorium hydroxide, and then through the testing vessel at a slow enough rate so that practically all the emanation would decay in the vessel. The activities of the rod and case, the rod being used as cathode, were measured by means of a Dolezalek electrometer. The activity of the rod was measured in a cylindrical vessel, free from active deposit, of the same dimensions as the anode case; while the latter was tested with a clean rod similar in dimensions to that used as cathode. The same general results were obtained as with the parallel plate vessels although the voltage necessary to bring over a given fraction of the rest atoms on to the cathode was higher owing to the difference in shape of the vessels. A similar rise of percentage cathode activity with voltage was found. With 280 volts (the highest used), the percentage cathode activity was 95.3.

From these experiments it will be seen that parallel plates and cylindrical vessels give concordant results.

To test if the variations between thorium and radium might be due to a surface effect of the electrodes, experiments were made to see if the surface conditions of the electrodes had any effect on the distribution of the rest atoms.

Experiments with copper and lead electrodes gave results similar to those obtained with zinc plates. The state of polish of the electrodes also had no effect on the distribution. These results are in accordance with those found for radium by Wellisch[1] and by Walmsley[2].

An interesting result found by Godlewski[3] might be referred to here. He found that on electrolyzing a solution of actinium, the active deposit was transported to the cathode, only when the cathode plate had previously been used as

1. Wellisch, Am. Journ. Sci. Oct. 13.
2. Walmsley Phil. Mag. Oct. 14.
3. Godlewski, Bull. Acad. Sci. de Cracovie, June 1913.

anode, and hence was electrolytically saturated with hydrogen. To test if a similar effect took place with thorium in air, experiments were made, using as cathodes plates previously used as anodes, and *vice versa*. No effect was detected within the limits of observational error.

It was found by Wellisch and Bronson that in the case of radium a much greater proportion of the rest atoms were uncharged in an atmosphere containing water vapor than in dry air. The effect of water vapor on the distribution of the rest atoms of thorium was accordingly tried. In order to obtain the maximum effect possible with water vapor experiments were made with air saturated with water vapor at room temperatures. This was accomplished by substituting water for the sulphuric acid of the previous experiments so that the plates stood over water. As the temperature of the laboratory varied from day to day the conditions under which the different results were obtained were not identical. The general effect however is seen in table 2, and is plotted in curve B, Fig. 4.

TABLE 2.

Volts.	Percentage cathode activity.
6.4	66.5
14.1	73.7
20.3	78.8
40.0	84.8
99.7	93.4
196.	96.0
313.	97.2

The effect of water vapor is evidently to greatly increase the rate of recombination. The question of its effect on the initial charged condition of the rest atoms will be referred to after the effects of ether vapor have been discussed.

It was found by Wellisch[1] that in an atmosphere of 8 cm. of ether vapor all the radium rest atoms were uncharged. The effect of adding ether vapor to the air was found to be very marked in the case of thorium. The apparatus was placed under a bell jar from which the air was then exhausted. Ether vapor in different quantities was added, air was then allowed to enter till atmospheric pressure was reached. The results obtained are shown in table 3 and graphically in fig. 5. The potential employed was 40 volts and old type vessels were used.

TABLE 3.

Ether added in cm. mercury.	Percentage cathode activity.
0	96.5
3.8	83.8
9.9	68.0
15.7	60.6
37.2[2]	54.5

The effect of different potentials in the case of ether vapor is shown in table 4. The ether vapor was saturated at room temperatures and the results therefore are not strictly comparable. The general effect however is seen. Old type vessels were used.

TABLE 4.

Volts.	Percentage cathode activity.
13.9	51.1
40.0	54.5
292.0	62.3

1. Wellisch, Phil. Mag, Oct. 1914.
2. Estimated, air saturated.

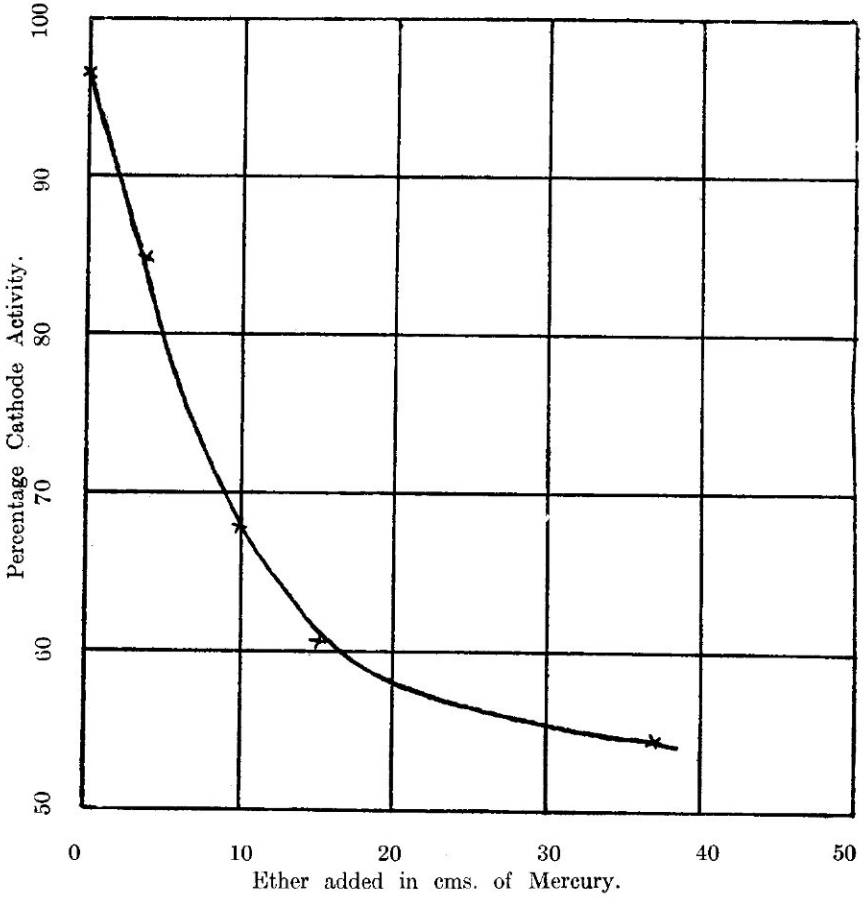


FIG. 5.

Two conclusions are suggested from these tables. 1st. With a given voltage the percentage cathode activity depends on the quantity of ether vapor present. 2nd. There is no evidence that with high potentials a value at all near 100 exists for the percentage cathode activity. This evidence points to the conclusion that part of the rest atoms are initially charged and part uncharged, and that the relative amounts of charged and uncharged rest atoms depend on the relative quantities of ether and air.

To further test this point, the following experiments were tried, the voltage in both cases being 313.

1st. The testing vessel was placed under a bell jar, the air exhausted and 10.2 cm. of ether vapor allowed to enter. Then air was admitted till atmospheric pressure was reached. The percentage cathode activity was 63.8.

2nd. The testing vessel was set up under a bell jar as before and the air exhausted to 0.7 cm. Then 10.2 cm. ether vapor were added and the exposure taken under a total pressure of 10.9 cm. The percentage cathode activity was 50.8.

A similar effect in the case of water vapor was sought. The bell jar containing a dish of water in addition to the testing vessel was exhausted to 1.6 cm. and an exposure taken using a potential of 312 volts. Owing to leakage the final pressure was 2.3 cm. The percentage cathode activity was 79.9. The exact proportions of water vapor and air in the bell jar were hard to determine but at least half the pressure was due to water vapor.

At such low pressures the percentage of the active deposit on the anode is increased due to recoil of the rest atoms. In order to test whether recoil could wholly account for the low value of the percentage cathode activity with water vapor, an exposure was taken using dry air at initial and final pressures of 1.35 and 1.55 cm. respectively. The percentage cathode activity was 94.6.

Thus we see that in an atmosphere of pure ether all the rest atoms are uncharged, which further strengthens the conclusions drawn from tables 3 and 4. The results with water vapor point towards similar conclusions, notwithstanding the experimental difficulty; the pressure of the water vapor being necessarily small, and there being a considerable fraction of air present.

Experiments tried in atmospheres of various other substances gave no evidence that in any pure atmosphere the

rest atoms were initially partly charged and partly uncharged. In this respect thorium seems to differ from radium. It was shown by Wellisch that, in hydrogen and in carbon dioxide, the rest atoms of the latter are partly charged and partly uncharged. The case of air is peculiar as it is a mixture. Further investigation along these lines is in progress.

Summary.

A new type of vessel for this kind of work has been developed and its advantages noted.

It has been shown that in dry air all the thorium rest atoms are initially positively charged.

In pure ether vapor all the thorium rest atoms are initially uncharged and in a mixture of ether vapor and air the charged condition of the rest atoms depends on the relative amounts of ether vapor and air present. A similar state of affairs appears to be true with water vapor.

In conclusion I wish to express my best thanks to Dr. Bronson, who suggested the work and without whose continued interest this work would not have been possible.