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I. *A Radio-active Substance emitted from Thorium Compounds.*

By E. RUTHERFORD, *M.A., B.Sc., Macdonald Professor of Physics, McGill University, Montreal* *.

IT has been shown by Schmidt† that thorium compounds give out a type of radiation similar in its photographic and electrical actions to uranium and Röntgen radiation. In addition to this ordinary radiation, I have found that thorium compounds continuously emit radio-active particles of some kind, which retain their radio-active powers for several minutes. This "emanation," as it will be termed for shortness, has the power of ionizing the gas in its neighbourhood and of passing through thin layers of metals, and, with great ease, through considerable thicknesses of paper.

In order to make clear the evidence of the existence of a radio-active emanation, an account will first be given of the anomalous behaviour of thorium compounds compared with those of uranium. Thorium oxide has been employed in most of the experiments, as it exhibits the "emanation" property to a greater degree than the other compounds; but what is true for the oxide is also true, but to a less extent, of the other thorium compounds examined, viz., the nitrate, sulphate, acetate, and oxalate.

In a previous paper‡ the author has shown that the radiation

* Communicated by Prof J. J. Thomson, F.R.S.

† Wied. *Ann.* May 1898.

‡ Phil. Mag. Jan. 1899, p. 109.

from thorium is of a more penetrating character than the radiation from uranium. Attention was also directed to the inconstancy of thorium as a source of radiation. Owens* has investigated in more detail the radiation from thorium compounds. He has shown that the radiations from the different compounds are of the same kind, and, with the exception of thorium oxide in thick layers, approximately homogeneous in character.

The intensity of thorium radiation, when examined by means of the electrical discharge produced, is found to be very variable; and this inconstancy is due to slow currents of air produced in an open room. When the apparatus is placed in a closed vessel, to do away with air-currents, the intensity is found to be practically constant. The sensitiveness of thorium oxide to slight currents of air is very remarkable. The movement of the air caused by the opening or closing of a door at the end of the room opposite to where the apparatus is placed, is often sufficient to considerably diminish the rate of discharge. In this respect thorium compounds differ from those of uranium, which are not appreciably affected by slight currents of air. Another anomaly that thorium compounds exhibit is the ease with which the radiation apparently passes through paper. The following table is an example of the way the rate of leak between two parallel plates, one of which is covered with a *thick* layer of thorium oxide, varies with the number of layers of ordinary foolscap-paper placed over the radio-active substance.

TABLE I.

Thickness of each Layer of Paper = ·008 cm. 50 volts between plates.

Number of Layers of Paper.	Rate of Discharge.
0	1
1	·74
2	·74
5	·72
10	·67
20	·55

* Phil. Mag. Oct. 1899, p. 360.

In the above table the rate of leak with the thorium oxide uncovered is taken as unity. It will be observed that the first layer reduced the rate of leak to ·74, and the five succeeding layers produce very little effect.

The action, however, is quite different if we use a *thin* * layer of thorium oxide. With one layer of paper, the rate of discharge is then reduced to less than $\frac{1}{16}$ of its value. At first sight it appears as if the thorium oxide gave out two types of radiation, one of which is readily absorbed by paper, and the other to only a slight extent. If we examine the radiation given out by a thin layer of thorium oxide, by placing successive layers of thin paper upon it, we find the radiation is approximately homogeneous, as the following table shows.

TABLE II.

Thickness of Paper = ·0027 cm.

Number of Layers of Thin Paper.	Rate of Discharge.
0	1
1	·37
2	·16
3	·08

The rate of leak of the bare salt is taken as unity. If the radiation is of one kind, we should expect the rate of discharge (which is proportional to the intensity of the radiation) to diminish in geometrical progression with the addition of equal thicknesses of paper. The above figures show that this is approximately the case. With a thick layer of thorium oxide, by adding successive layers of thin paper, we find the rate of discharge gradually diminish, till after a few layers it reaches a constant value. The amount that is cut off by the first layer of foolscap-paper (see Table I.) is of the same kind of radiation as that which is emitted by a thin layer of oxide.

On directing a slight current of air between the test-plates, the rate of discharge due to a thick layer of thorium oxide is

* To produce a thin layer on a plate, the oxide, in the form of a fine powder, was sprinkled by means of a fine gauze, so as to cover the plate to a very small depth. By a thick layer is meant a layer of oxide over a millimetre in thickness.

greatly diminished. The amount of diminution is to a great extent independent of the electromotive force acting between the plates. Under similar conditions with uranium, the rate of leak is not appreciably affected. With a thin layer of oxide, the diminution of the rate of leak is small; but with a thick layer of oxide, the rate of leak may be reduced to less than one-third of its previous value. If two thicknesses of foolscap-paper are placed over the thorium oxide, the resulting rate of leak between the plates may be diminished to less than $\frac{1}{20}$ of its value by a slight continuous blast of air from a gasometer or bellows.

The phenomena exhibited by thorium compounds receive a complete explanation if we suppose that, in addition to the ordinary radiation, a large number of radio-active particles are given out from the mass of the active substance. This "emanation" can pass through considerable thicknesses of paper. The radio-active particles emitted by the thorium compounds gradually diffuse through the gas in its neighbourhood and become centres of ionization throughout the gas. The fact that the effect of air-currents is only observed to a slight extent with thin layers of thorium oxide is due to the preponderance, in that case, of the rate of leak due to the ordinary radiation over that due to the emanation. With a thick layer of thorium oxide, the rate of leak due to the ordinary radiation is practically that due to a thin surface-layer, as the radiation can only penetrate a short distance through the salt. On the other hand, the "emanation" is able to diffuse from a distance of several millimetres below the surface of the compound, and the rate of leak due to it becomes much greater than that due to the radiation alone.

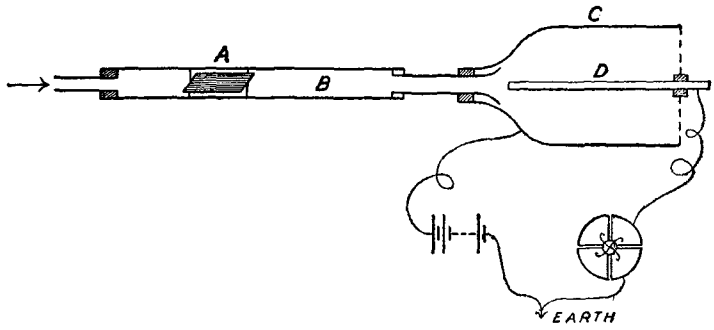
The explanation of the action of slight currents of air is clear on the "emanation" theory. Since the radio-active particles are not affected by an electrical field, extremely minute motions of air, if continuous, remove many of the radio-active centres from between the plates. It will be shown shortly that the emanation continues to ionize the gas in its neighbourhood for several minutes, so that the removal of the particles from between the plates diminishes the rate of discharge between the plates.

Duration of the Radio-activity of the Emanation.

The emanation gradually loses its radio-active power. The following method was adopted to determine the rate of decay of the intensity of the radiation of the radio-active particles emitted by thorium oxide.

A thick layer of thorium oxide was enclosed in a narrow rectangular paper vessel A (fig. 1), made up of two thicknesses of foolscap-paper. The paper cut off the regular radiation almost entirely, but allowed the emanation to pass through. The thorium thus enclosed was placed inside a

Fig. 1.



long metal tube B. One end of the tube was connected to a large insulated cylindrical vessel C, which had a number of small holes in the end for the passage of air. Inside C was fixed an insulated electrode, D, connected with one pair of quadrants of a Thomson electrometer. The cylinder, C, was connected to one terminal of a battery of 100 volts, the other terminal of which was connected to earth.

A slow current of air from an aspirator or gasometer, which had been freed from dust by its passage through a plug of cotton-wool, was passed through the apparatus. The current of air, in its passage by the thorium oxide, carried away the radio-active particles with it, and these were gradually conveyed into the large cylinder C. The electrometer-needle showed no sign of movement until the radio-active particles were carried into C. In consequence of the ionization of the gas in the cylinder by the radio-active particles, a current passed between the electrodes C and D. The value of the current was the same whether C was connected with the positive or negative pole of the battery. When the current of air had been flowing for some minutes, the current between C and D reached a constant value. The flow of air was then stopped, and the rate of leak between C and D observed at regular intervals. It was found that the current between C and D persisted for over ten minutes.

The following is a series of observations.

TABLE III.

Potential-difference 100 volts.

Time in Seconds.	Current.
0	1
28	·69
62	·51
118	·23
155	·14
210	·067
272	·041
360	·018

Fig. 2.

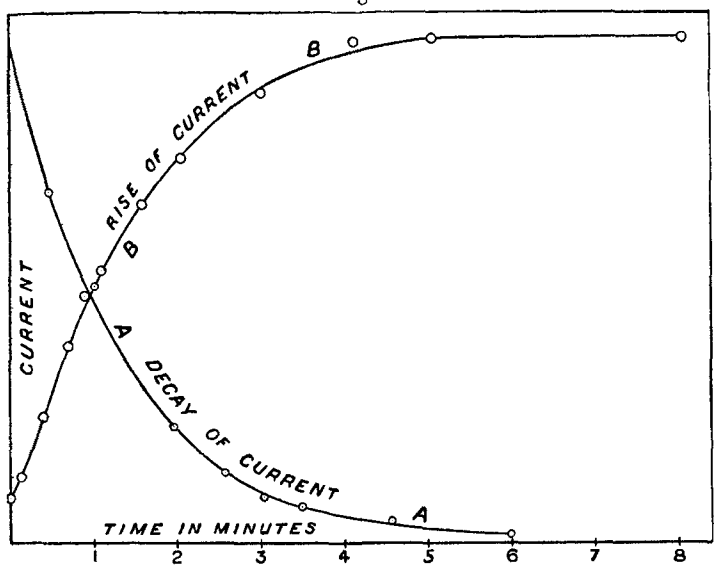


Fig. 2, curve A, shows the relation existing between the current through the gas and the time. The current, just before the flow of air is stopped, is taken as unity. It will be observed that the current through the gas diminishes in a geometrical progression with the time. It can easily be shown,

by the theory of ionization, that the current through the gas is proportional to the intensity of the radiation emitted by the radio-active particles. We therefore see that the intensity of the radiation given out by the radio-active particles falls off in a geometrical progression with the time. The result shows that the intensity of the radiation has fallen to one-half its value after an interval of about *one minute*. The rate of leak due to the emanation was too small for measurement after an interval of ten minutes.

If the ionized gas had been produced from a uranium compound, the duration of the conductivity, for voltages such as were used, would only have been a fraction of a second.

The rate of decay of intensity is independent of the electromotive force acting on the gas. This shows that the radio-active particles are not destroyed by the electric field. The current through the gas at any particular instant, after stoppage of the flow of air, was found to be the same whether the electromotive force had been acting the whole time or just applied for the time of the test.

The current through the gas in the cylinder depends on the electromotive force in the same way as the current through a gas made conducting by Röntgen rays. The current at first increases nearly in proportion to the electromotive force, but soon reaches an approximate "saturation" value.

The duration of the radio-activity was also tested by another method. The paper vessel containing the thorium oxide was placed inside a long brass cylinder over 200 cms. in length. A slow current of air (with a velocity of about 2 cms. per second along the tube) was passed over the thorium oxide along the tube, and then between two insulated concentric cylinders. The rate of leak between the two concentric cylinders (potential-difference 270 volts) was observed when the air had been passing sufficiently long to produce a steady state. The rates of leak were observed for varying positions of the thorium oxide along the tube. Knowing the velocity of the current of air along the tube, the time taken to carry the radio-active particles to the testing-apparatus could be determined. In this way it was found that the rate of decay was about the same as determined by the first method, *i. e.*, the intensity fell to half its value in about one minute.

In this apparatus experiments were also tried to see whether the radio-active particles moved in an electric field. The experiments on the effect of a current of air on the rate of discharge naturally suggest that possibly one of the ions was so large that it moved extremely slowly even in strong electric fields. The results obtained showed that the particles did not move

with a greater velocity than $\frac{1}{100,000}$ cm. per second for a potential-gradient of one volt per cm.; and it is probable that the particles do not move at all in an electric field. By blowing the emanation into an inductor, no evidence of any charge in the emanation could be detected. We may therefore conclude that the emanation is uncharged, and is not appreciably affected by an electric field.

Properties of the Emanation.

The emanation passes through a plug of cotton-wool without any loss of its radio-active powers. It is also unaffected by bubbling through hot or cold water, weak or strong sulphuric acid. In this respect it acts like an ordinary gas. An ion, on the other hand, is not able to pass through a plug of cotton-wool, or to bubble through water, without losing its charge.

The emanation is similar to uranium in its photographic and electrical actions. It can ionize the gas in its neighbourhood, and can affect a photographic plate in the dark after several days' exposure. Russell * has shown that the active agent in producing photographic action in the case of metals, paper, &c., is due to hydrogen peroxide. Hydrogen peroxide apparently has the power of passing in some way through considerable thicknesses of special substances, and in this respect the emanation resembles it. Hydrogen peroxide, however, does not ionize the gas in its neighbourhood. The action of hydrogen peroxide on the photographic plate is purely a chemical one; but it is the radiation from the emanation, and not the emanation itself, that produces ionizing and photographic actions.

The radio-active emanation passes through all metals if sufficiently thin. In order to make certain that the emanation passed through the material to be examined and did not diffuse round the edges, the radio-active substance was placed in a square groove of a thick lead plate. Two layers of paper were pasted tightly over the opening to cut off the regular radiation. The material to be tested was then firmly waxed down on the lead plate.

The following numbers illustrate the effect of different metals. The rate of discharge, due to the emanation between two parallel plates 4 cms. apart, was observed.

* Proc. Roy. Soc. 1897.

Aluminium Foil, thickness = ·0008 cm.

Number of Layers.	Rate of Discharge.
0	1
1	·66
3	·42
6	·16

Cardboard, thickness ·08 cm.

Layers.	Rate of Discharge.
0	1
1	·40
2	·21

The emanation passed readily through several thicknesses of gold- and silver-leaf. A plate of mica, thickness ·006 cm., was completely impervious to the emanation.

When a thick layer of thorium oxide, covered over with several thicknesses of paper, is placed inside a closed vessel, the rate of discharge due to the emanation is small at first, but gradually increases, until after a few minutes a steady state is reached.

These results are to be expected, for the emanation can only slowly diffuse through the paper and the surrounding air. A steady state is reached when the rate of loss of intensity due to the gradual decay of the radio-activity of the emanation is recompensed by the number of new radio-active centres supplied from the thorium compound.

Let n = number of ions produced per second by the radio-active particles between the plates.

Let q = number of ions supplied per second by the emanation diffusing from the thorium.

The rate of variation of the number of ions at any time t is given by

$$\frac{dn}{dt} = q - \lambda n,$$

where λ is a constant.

The results given in Table III. show that the rate of diminution of the number of ions is proportional to the number present.

Solving the equation, it is seen that

$$\log_e (q - \lambda n) = -\lambda t + A,$$

where A is a constant.

When $t=0, \quad n=0;$

therefore $A = \log_e q.$

Thus

$$n = \frac{q}{\lambda}(1 - e^{-\lambda t}).$$

With a large potential-difference between the test-plates the current i through the gas at any time is given by

$$i = ne,$$

where e is the charge on an ion.

When a steady state is reached, $\frac{dn}{dt} = 0$; and the maximum number N of ions produced per second by the radio-active particles between the plates is given by

$$N = \frac{q}{\lambda},$$

and the maximum current I is given by

$$I = Ne.$$

Therefore

$$\frac{i}{I} = 1 - e^{-\lambda t}.$$

The current thus increases according to the same law as a current of electricity rises in a circuit of constant inductance.

This result is confirmed by an experiment on the rise of the current between two concentric cylinders. The thorium oxide enclosed in paper was placed inside the cylinder. A current of air was sent between the cylinders in order to remove the emanation as rapidly as it was formed. The current of air was then stopped and the current between the two cylinders observed, by means of an electrometer, for successive intervals after the current of air ceased. Table IV. gives the results obtained.

TABLE IV.

Length of cylinder = 30 cms.

Internal diameter outer cylinder = 5·5 cms.
External „ inner „ = ·8 cm.

100 volts between cylinders.

Time in Seconds.	Current in Scale-divisions per second.
0	2·4
7·5	3·3
2 3	6·5
4 0	10·0
5·3	12·5
6·7	13·8
9·6	17·1
12·5	19·4
18·4	22·7
24·4	25·3
30 4	25·6
48·4	25·6

The results are expressed in fig. 2, curve B, where the ordinate represents current and the abscissa time. It will be observed that the curve of rise of the current is similar in form to the rise of an electric current in a circuit of constant inductance. The current reaches half its value about one minute after the current of air has stopped,—a result which agrees with the equation given, for $e^{-\lambda t} = \frac{1}{2}$ when $t = 60$ seconds (see Table IV.). At the instant of stopping the current of air the current has a definite value, since most of the ions given off by the emanation, before it is blown out of the cylinders, reach the electrodes.

When the source of the emanation is removed, $q = 0$, and the decay of the number of ions produced by the emanation is given by the equation

$$\frac{dn}{dt} = -\lambda n.$$

If $n = N$ when $t = 0$, it is easily seen that

$$\frac{n}{N} = e^{-\lambda t},$$

or

$$\frac{i}{I} = e^{-\lambda t};$$

i. e., the current through the gas diminishes in a geometrical progression. After 20 minutes the current through the gas is only about one millionth part of its initial value.

It has been shown that $e^{-\lambda t} = \frac{1}{2}$ when $t = 60$ seconds.

Therefore $\lambda = \frac{1}{86}$,

and

$$N = \frac{q}{\lambda} = 86\,q;$$

or the total number of ions produced per second when a steady state is reached is 86 times the number of ions supplied per second by the emanation.

The amount of emanation from thorium oxide increases with the thickness of the layer. When 1 gramme of thorium oxide was spread over a surface of 25 cms., the amount of discharge due to the ordinary radiation had practically reached a maximum. The rate of leak due to the emanation for the same thickness was small. With 9 grammes of oxide spread over the same area, the rate of leak due to the emanation had reached about half its maximum value, which for that case corresponded to four times the rate of leak caused by the ordinary radiation. The emanation thus still preserves its radio-active properties after diffusing through several millimetres of thorium compound.

The emanation is given out whatever the gas by which the thorium is surrounded. The action is very similar whether air, oxygen, hydrogen, or carbonic acid is used.

The rate of discharge due to the emanation diminishes with lowering of the pressure of the air surrounding it. Only a few observations have been made, but the results seem to point to a uniform rate of emission of the emanation at all pressures; but since the intensity of the ionization of the gas varies directly as the pressure, the rate of leak decreases with lowering of the pressure.

The amount of the emanation, so far as the experiments have gone, is also independent of the quantity of water-vapour present.

The power of emitting radio-active particles is not possessed to any appreciable extent by other radio-active substances besides thorium. All the compounds of thorium examined possess it to a marked degree, and it is especially large in the oxide. Two different specimens of the oxide have been used, one obtained from Schuchart of Germany, and the

other from Eimer & Amend of New York. The oxide is prepared by the latter by igniting thorium nitrate obtained from monazite sand.

The amount of discharge caused by the emanation is increased several times by the conversion of the nitrate into the oxide; but at the same time, the rate of discharge due to the ordinary radiation emitted by the thorium is increased in about an equal ratio. The conversion of the nitrate into the oxide took place below a red heat. On heating in a muffle for some time at white heat, the amount of emanation continually diminished, till after four hours' exposure to the heat, the rate of discharge due to the emanation was only $\frac{1}{20}$ of the value immediately after its conversion into oxide.

Both thorium oxalate and sulphate act in a similar manner to the nitrate; but the emanation is still given off to a considerable extent after continued heating.

In considering the question of the origin and nature of the emanation, two possible explanations naturally suggest themselves, viz.:—

(1) That the emanation may be due to fine dust particles of the radio-active substance emitted by the thorium compounds.

(2) That the emanation may be a vapour given off from thorium compounds.

The fact that the emanation can pass through metals and large thicknesses of paper and through plugs of cotton-wool, is strong evidence against the dust hypothesis. Special experiments, however, were tried to settle the question. The experiments of Aitken and Wilson * have shown that ordinary air can be completely freed from dust particles by repeated small expansions of the air over a water-surface. The dust particles act as nuclei for the formation of small drops, and are removed from the gas by the action of gravity.

The experiment was repeated with thorium oxide present in the vessel. The oxide was enclosed in a paper cylinder, which allowed the emanation to pass through it. After repeated expansions no cloud was formed, showing that for the expansions used the particles of the emanation were too small to become centres of condensation of the water-vapour. We may therefore conclude, from this experiment, that the emanation does not consist of dust particles of thorium oxide.

It would be of interest to examine the behaviour of the emanation for greater and more sudden expansions, after the

* Trans. Roy. Soc. 1897.

manner employed by C. T. R. Wilson* in his experiments on the action of ions as centres of condensation.

The emanation may possibly be a vapour of thorium. There is reason to believe that all metals and substances give off vapour to some degree. If the radio-active power of thorium is possessed by the molecules of the substance, it would be expected that the vapour of the substance would be itself radio-active for a short time, but the radio-active power would diminish in consequence of the rapid radiation of energy. Some information on this point could probably be obtained by observation of the rate of diffusion of the emanation into gases. It is hoped that experimental data of this kind will lead to an approximate determination of the molecular weight of the emanation.

Experiments have been tried to see if the amount of the emanation from thorium oxide is sufficient to appreciably alter the pressure of the gas in an exhausted tube. The oxide was placed in a bulb connected with a Plücker spectroscopic tube. The whole was exhausted, and the pressure noted by a McLeod gauge. The bulb of thorium oxide was disconnected from the main tube by means of a stopcock. The Plücker tube was refilled and exhausted again to the same pressure. On connecting the two tubes together again, no appreciable difference in the pressure or in the appearance of the discharge from an induction-coil was observed. The spectrum of the gas was unchanged.

Experiments, which are still in progress, show that the emanation possesses a very remarkable property. I have found that the positive ion produced in a gas by the emanation possesses the power of producing radio-activity in all substances on which it falls. This power of giving forth a radiation lasts for several days. The radiation is of a more penetrating character than that given out by thorium or uranium. The emanation from thorium compounds thus has properties which the thorium itself does not possess. A more complete account of the results obtained is reserved for a later communication.

McGill University, Montreal,
September 13th, 1899.

* Phil. Trans. Roy. Soc. vol. clxxxix. (1897).