THE

LONDON, EDINBURGH, AND DUBLIN

PHILOSOPHICAL MAGAZINE

AND

JOURNAL OF SCIENCE.

[FIFTH SERIES.]

FEBRUARY 1900.

XI. *Radioactivity produced in Substances by the Action of Thorium Compounds.* *By* E. RUTHERFORD, *M.A., B.Sc., Macdonald Professor of Physics, McGill University, Montreal* *.

THORIUM compounds under certain conditions possess the property of producing temporary radioactivity in all solid substances in their neighbourhood. The substance made radio-active behaves, with regard to its photographic and electrical actions, as if it were covered with a layer of radio-active substance like uranium or thorium. Unlike the radiations from thorium and uranium, which are given out uniformly for long periods of time, the intensity of the excited radiation is not constant, but gradually diminishes. The intensity falls to half its value about eleven hours after the removal of the substance from the neighbourhood of the thorium. The radiation given out is more penetrating in character than the similar radiations emitted by uranium and thorium and the radio-active derivatives from pitchblende, radium †, and polonium ‡.

Attention was first drawn to this phenomenon of what may be termed "excited radioactivity" by the apparent failure of good insulators, like ebonite and paraffin, to continue to insulate in the presence of thorium compounds.

The apparatus first used is shown in fig. 1.

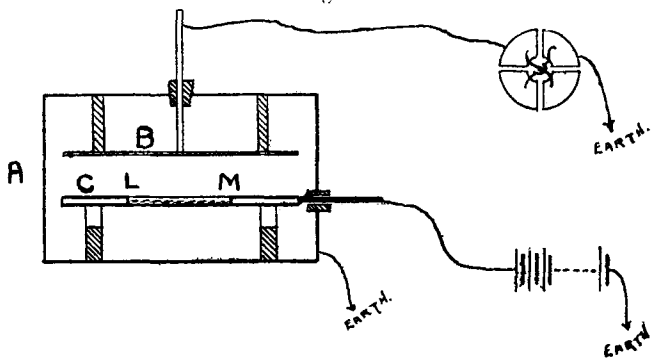
Two insulated plates, B and C, were placed parallel to one another. In a shallow square depression LM in the plate C,

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

† Curie, *C. R.* 1898, p. 175. ‡ Curie, *ibid.* 26 Dec. 1898

a layer of thorium oxide was placed and covered with several layers of foolscap-paper. The whole was enclosed in a lead vessel A, with a door in the side to allow the plate C to be

Fig. 1.



readily moved. The crossed lines show the position of insulators. The plate C was connected to the + pole of a battery of 50 volts, the other terminal of which was to earth. The plate B was connected to one pair of quadrants of a delicate Thomson electrometer with a replenisher and gauge, the other pair of quadrants of which was connected to earth.

With the arrangement in the figure, when B is insulated, there can be no conduction-current from C along or through the insulators, since the earth-connected vessel intervenes. If the thorium-covered plate C was removed, and a brass one of the same dimensions substituted, there was no appreciable movement of the electrometer-needle. If, however, the plate C, covered with thorium oxide, were left in the vessel for several hours with the plate B charged —, on removal of C and the substitution of a non-active metal plate, the movement of the electrometer-needle showed that B was receiving a + charge. On reversing the battery, the current was reversed in direction but equal in amount. The current between the plates gradually decreased with the time, and became inappreciable after a few days. By replacing the thorium oxide, the experiment could be repeated.

It was at first thought that possibly dust particles from the thorium oxide might have escaped from under the paper and in some way adhered to the upper plate. An examination of the plate B, however, revealed no trace of thorium oxide on its surface. The plate made the air a conductor in its neighbourhood, as if it were covered with a thick layer of radio-active substance. If the surface of the plate was care-

fully scrubbed with sand- or emery-paper, the radio-active power was to a great extent destroyed. It was found possible to make the plate B active, even if the thorium oxide were covered with 30 layers of foolscap paper tightly waxed down so as to prevent the escape of dust particles.

If the plate C was charged — and B+, the plate B no longer became radio-active, but the top layer of paper over the thorium was found to be active on its upper side to about the same extent as the plate B in the previous case; *i.e.*, the negatively charged surface was made active in both cases.

All the compounds of thorium examined have the power of causing radioactivity in substances. The oxide, however, gives far the largest effects, and has consequently been used in most of the experiments.

The thorium compounds used were supplied by Messrs. Eimer & Amend, New York. The oxide was obtained by igniting the nitrate which had been manufactured from monazite sand. If the oxide is heated for some hours to a white heat in a platinum crucible, it loses its power of exciting radioactivity in substances to a very large extent.

Comparison of Intensities of Radiation.

The intensity of the radiation, excited in substances in the manner described, was in all cases compared by the electrical method. In general, for the purposes of measurement, the radioactivity was excited in flat plates or circular cylinders.

For flat plates the testing apparatus was similar to fig. 1. The brass plates corresponding to B and C were 5 cms. apart, with a potential-difference of 50 volts between them. The current between the plates, measured by the rate of movement of the electrometer-needle, was taken as proportional to the intensity of the radiation at the surface. With radio-active cylinders, the active cylinder was placed in a larger cylinder and concentric with it. The current for 50 volts between the cylinders was taken as a measure of the intensity of the radiation at the surface.

For experiments, extending in some cases over several days or weeks, it was necessary that for each observation the electrometer should be of the same degree of sensitiveness. This was roughly ensured by the Thomson Replenisher and Gauge, attached to the electrometer. For small variations from the standard sensitiveness, the values of the current were corrected by observing the number of divisions on the electrometer-scale corresponding to the E M.F. of a Clark cell.

As in the course of this paper it will be necessary to compare

the intensity of the radiation from radio-active plates and cylinders, a brief theoretical discussion will be given of the relation that exists between the intensity of the radiation, the area of the active surface, and the maximum current through the gas.

Two cases will be considered—

(I.) When the radiation is given out uniformly from a plane surface and the current through the gas is measured between two parallel planes.

(II.) When the radiation is given out from a cylinder and the current measured between concentric cylinders.

Case I.—We will first consider the case of a uniformly radio-active plate C, of area S, which is placed between two large parallel plates A and B (fig. 2 *a*).

Fig. 2 *a*.

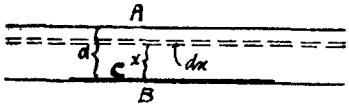
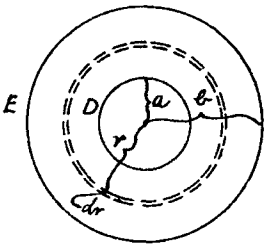


Fig. 2 *b*.



We will suppose the plate C to be of large dimensions compared with the distance d of the plate C from A, and to give out radiation equally from all points of its surface. The gas is ionized by the passage of the radiation through it, and the ions produced travel to the plates A and C under the influence of the electric field.

In consequence of the energy required to ionize the gas, the intensity of the radiation diminishes in its passage through it.

Suppose the radiation is homogeneous in character, and that λ is the coefficient of absorption of the radiation by the gas. Let I_0 be the intensity of the radiation at the surface of the plate. Since the plate is large compared with the distance d, the value of the intensity may be considered approximately equal at equal distances from the surface C. In consequence of the absorption of the radiation by the gas, the intensity I at a distance x from the active plate is given by

$$I = I_0 e^{-\lambda x}.$$

Let dn be the number of ions produced per second between two planes parallel to C and distant x and $x+dx$ from it.

Since the rate of production of ions is proportional to the

intensity of the radiation, the total number of ions n produced per second between A and C, distant d apart, is given by

$$n = \int_0^d \mathrm{KSI}_0 e^{-\lambda x} dx, \text{ where K is a constant,}$$
$$= \frac{\mathrm{KSI}_0}{\lambda}(1-e^{-\lambda d}).$$

If ϵ is the charge on an ion, the current i through the gas, when an E.M.F. is applied sufficient to remove all the ions before recombination takes place, is given by

$$i = n\epsilon.$$

Therefore

$$\mathrm{SI}_0 = \frac{i\lambda}{\mathrm{K}\epsilon(1-e^{-\lambda d})};$$

or the product of the intensity of the radiation and the area of the active surface is proportional to the current through the gas.

It is of interest to develop the above equation from considerations of the energy required to produce an ion.

Let W be the average amount of energy used up in producing an ion in the gas. We will assume that the absorption of the energy of the radiation in its passage through the gas is due solely to the production of ions. On account of the absorption, the intensity of the radiation varies from I_0 at the surface of the active plate to $\mathrm{I}_0 e^{-\lambda d}$ at the surface of the top plate. If n be the total number of ions produced, we thus obtain

$$n.\mathrm{W} = \mathrm{SI}_0(1-e^{-\lambda d}),$$

where the energy absorbed over an area S is given by the right-hand side of the equation; or

$$\mathrm{SI}_0 = \frac{\mathrm{W}i}{\epsilon(1-e^{-\lambda d})},$$

where current $i = n\epsilon$, as before.

Some experiments given in previous papers* point to the conclusion that the energy required to produce an ion may possibly be the same for all gases at all pressures, and it has been shown by Prof. J. J. Thomson and Mr. Townsend that the charge of the ions† in different gases is the same. If such is the case, W/ϵ is a constant for all gases and the current through the gas will depend only on λ, d, and SI_0.

* Rutherford, Phil. Mag. Jan. 1899.

† J. J. Thomson, Phil. Mag. Dec. 1898; J. S. Townsend, Trans. Roy. Soc. 1899.

Case II.—We will now consider the case of a radio-active cylinder, where the current is measured between two concentric cylinders. Let fig. 2 *b* represent a cross-section of the cylinders. Let a = radius of radio-active cylinder D, b = radius of concentric cylinder E. Suppose length of cylinder D to be large compared with the distance between the cylinders. If λ is the coefficient of absorption of the radiation, the intensity I at a distance r (outside D) from the centre is easily seen to be

$$\frac{I}{I_0} = \frac{a}{r} e^{-\lambda(r-a)},$$

where I_0 = intensity of radiation at the surface, since without any absorption the value of I would fall off inversely as the distance. The total energy of the radiation near the surface of the external cylinder is given per unit length by

$$I_0 \frac{a}{b} e^{-\lambda(b-a)} . 2\pi b,$$

the energy per unit length close to the surface of the active cylinder by $I_0 . 2\pi a$.

The total energy absorbed in the gas is thus equal to

$$I_0 . 2\pi a \{1 - e^{-\lambda(b-a)}\}.$$

If n = the number of ions produced per second due to the length l of the active rod,

$$W . n = I_0 . 2\pi a l \{1 - e^{-\lambda(b-a)}\}$$
$$= I_0 . S \{1 - e^{-\lambda(b-a)}\},$$

where S is surface-area of active cylinder;

or
$$SI_0 = \frac{W . i}{\epsilon \{1 - e^{-\lambda(b-a)}\}}, \text{ where } i = n\epsilon,$$
$$= \frac{Ai}{1 - e^{-\lambda(b-a)}}, \text{ where } A = \frac{W}{\epsilon} = \text{constant}.$$

In both of the cases considered, half the radiation has been absorbed in the substance which is made radio-active, and the other half passes through the gas, since the radiation is given out from the surface in all directions. In the case of complete absorption of the radiation in the passage through the gas, the maximum current i is given by

$$SI_0 = Ai.$$

An investigation is now in progress to determine the value of A, that is, W/ϵ. If A is determined, the intensity of the radiation can at once be expressed in absolute measure.

Conditions for the Production of Radioactivity in Substances.

In order to *confine* the induced radioactivity produced by thorium compounds to any particular conductor, it is necessary that it should be charged — and all other bodies in the field +. In order to produce radioactivity in all bodies in the neighbourhood, no electric field is required. If thorium oxide is placed in a closed vessel connected to earth, the sides of the vessel and any solid bodies near, whether conductors or insulators, become radio-active. If, in addition, the surface of the thorium oxide is covered with paper or thin aluminium-foil, the side of the paper away from the oxide becomes radio-active. When no electromotive forces are acting, the amount of radioactivity in a given time per unit area is greater the nearer the body to the thorium oxide.

With electromotive forces acting, the substance to which the radioactivity is due appears to travel along the lines of force from the + to the — charged body. It is thus possible to *concentrate* the radioactivity on small plates or fine wires by placing them in a closed metal vessel connected to earth and charging them —.

If the bodies are all uncharged, the particles producing radioactivity, by the process of diffusion through the gas, are carried to the sides of the bodies and adhere to them. A fine wire fixed in the centre of a vessel on the bottom of which the active salt is placed becomes only slightly radio-active, since only a few of the active particles reach its surface. The closer a body is to the thorium, other conditions remaining unaltered, the more active it becomes.

Fig. 3.

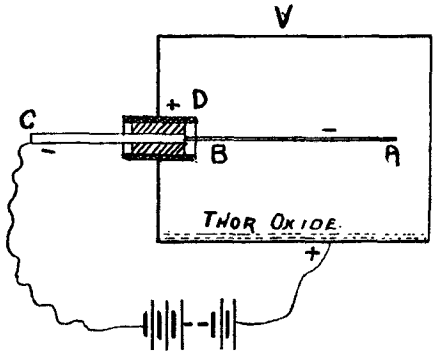


Fig. 3 shows the general arrangement for concentrating the activity on a small area of a conductor. A metal vessel V

was connected to the + pole of a battery of small lead accumulators of 300 volts, the other pole of which was to earth.

A thick layer of thorium oxide was placed in the bottom of the vessel and covered with several thicknesses of paper. A brass tube D was fixed in the side of the vessel and metallically connected with it. A fine platinum wire AB was fixed on the end of a stouter brass rod BC. The brass rod was fixed centrally in the cylinder D and insulated from it. The end of the brass rod B was placed well inside the cylinder D. The conductor AC was connected to earth.

The fine wire is thus the only body exposed in the field with a charge, and, under the influence of electric forces, the active particles are carried to the wire AB and adhere to its surface.

The same general results are obtained whether the surface of the thorium oxide is bare or covered with paper or thin layers of metal-foil.

Two or three layers of paper almost completely cut off the ordinary radiation * from thorium; so the effect cannot be due to the direct radiation from its surface.

In this way I have been able to cause a piece of platinum wire of length 1 cm. and diameter ·018 cm., *i. e.* with a surface area of ·056 cm., to give more than 20 times the rate of discharge given by a thick layer of uranium oxide of 25 sq. cms. area. A rate of movement of an electrometer-needle of 200 divisions in 5 seconds is quite easily obtained from the action of such a small active surface. (1 volt gave a deflexion of 40 divisions on the electrometer-scale, and the capacity of the whole circuit was about 50 electrostatic units.)

I have spoken of using a platinum wire, but any other metal wire will serve equally well. Using large electromotive forces and a large surface of thorium oxide, it would be quite possible to increase the radioactivity of unit area of the conductor to more than 20 times the value cited in the above case. So far as the results obtained indicate, there is no limit to the amount of increase, since we can suppose the area of the — charged conductor diminished and the amount of thorium increased. In practice, however, a limit would soon be reached, as it would be difficult to cause all the radio-active particles to move to the small conductor without very large electric forces.

* E. Rutherford, Phil. Mag. Jan. 1900.

Connexion between the "Emanation" from Thorium and "Excited" Radio-activity.

In a previous paper* I have shown that compounds of thorium emit some kind of radio-active material or "emanation," which is able to pass through considerable thicknesses of paper and thin layers of metal, and preserves its radiating power for several minutes. These particles diffuse through the gas and become centres of ionization throughout the volume of the gas. The current passing between two charged plates, on one of which is spread thorium oxide, is greatly diminished by directing a slow continuous blast of air between the plates. As the particles have no charge, they may be readily removed from between the plates by a current of air even in a strong electric field.

There is a very close connexion between this "emanation" and excited radioactivity—in fact, the emanation is in some way the direct cause of the latter. The following facts will serve to show the close connexion that exists :—

(1) All thorium compounds examined are able to make substances radio-active, but to different degrees. The greater the amount of emanation, the greater the amount of induced radioactivity. As an example, thorium oxide is the most active of all thorium compounds in producing radioactivity and giving out the emanation. A thin layer of thorium oxide gives out very little emanation, and is only slightly effective in producing radioactivity.

(2) Substances are made radio-active when the active compound is covered with several layers of paper or thin metal foil. The emanation also readily passes through paper and thin metal foil. Two or three layers of ordinary foolscap-paper completely cut off the ordinary radiation given out by thorium compounds, but do not much diminish the amount of induced radioactivity.

(3) A slow current of air, which quickly removes the emanation as it appears, also diminishes the power of producing radioactivity. The amount of induced radiation is greater in closed than in open vessels, on account of the disturbance of air-currents in the latter case.

(4) Thorium oxide which had been heated to a sufficiently high temperature gave out very little emanation and produced little radioactivity.

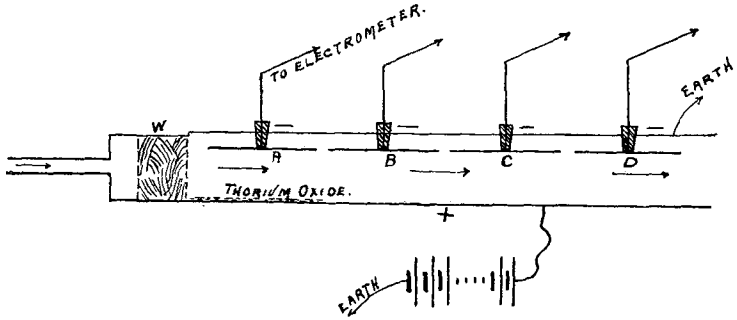
Speaking generally, it may be said that the presence of the emanation is necessary for the production of radioactivity in

* Phil. Mag. January 1900.

substances, and that the amount of radioactivity depends upon the amount of the "emanation." A radio-active substance like uranium, which gives out no emanation, produces no trace of excited radioactivity.

An experiment now to be described throws a further light on the question. The general arrangement of the experiment is shown in fig. 4.

Fig. 4.



A slow current of air from a gas-bag, after bubbling through sulphuric acid, passed down through a rectangular wooden vessel, 60 cms. in length. In order to remove spray and dust and to equalize the current of air over the cross-section, the air was passed through cotton-wool at W. A metal plate covered the bottom of the vessel and was charged +. Four insulated metal plates, A, B, C, D, placed at equal distances, were attached to a top metal plate connected to earth. Thorium oxide covered with paper was placed under the electrode A.

The current of air was passed through the vessel at the steady rate of about ·2 cm. per second for a period of 7 hours, with 300 volts between the lower and upper plates. The following results were obtained for the current due to the emanation which reached A, B, C, D and the corresponding radioactivity produced:—

	Relative current due to emanation.	Relative excited radioactivity.
Plate A . .	1	1
„ B . .	·55	·43
„ C . .	·18	·16
„ D . .	·072	·061

The current due to the emanation which reaches A, and the radioactivity produced in A, is in each case taken as unity for

the purpose of comparison. It will be observed that radio-activity is produced on the plates some distance away from the thorium oxide, and is roughly proportional to the emanation-current at the plate. We may conclude from this experiment that the radioactivity is, in some way, due to the "emanation," or to something that accompanies it, but is not caused by the direct action of a radiation from thorium oxide.

Absorption of the Radiation by Substances.

All radio-active substances, as well as bodies made radio-active in the manner described, ionize the gas in their neighbourhood and act upon a photographic plate in the dark. A simple method of testing whether two types of radiation are the same, is to determine the absorption of the radiation by layers of thin metal foil. If the absorption is different for the two types of radiation, we may consider them distinct kinds of radiation.

The current between two parallel plates, one surface of which was radio-active, was determined when successive layers of a substance of equal thickness were placed over the radio-active plate. The following table is an example of the way the current (which is proportional to the intensity of the radiation) diminishes with successive layers of aluminium-foil over a plate of zinc, which had been made radio-active:—

Zinc plate = 12 × 18 cms.
Thickness of foil = ·0004 cm.
95 volts between plates.

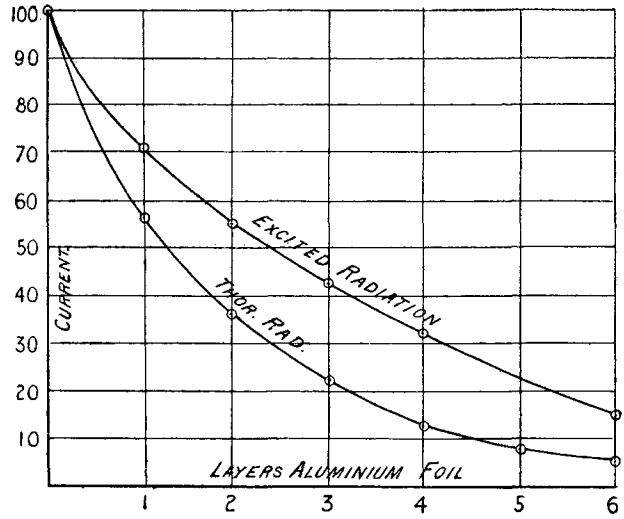
No. of layers alum. foil.	Current for radiation from zinc.	Current for thin layer of thorium oxide.
0	1	1
1	·71	·57
2	·55	·36
3	·43	·23
4	·32	·13
5	...	·084
6	·155	·056

The third column of the table gives the variation of the current with thickness of foil for a *thin* layer of thorium oxide, and serves as a basis of comparison with the excited radiation. The current for the bare radio-active surface is in each case taken as unity for the purpose of comparison.

Fig. 5, curves A, B, show these results graphically when the ordinates denote current and the abscissæ thicknesses of aluminium.

It will be observed that the radiations from zinc and thorium oxide are quite different in character, the radiation from the former being far more penetrating as regards aluminium. Both types of radiation are approximately homogeneous. The current, which is proportional to the intensity of the radiation, diminishes approximately in a geometrical progression as the thickness of the metal increases in arithmetical progression.

Fig. 5.



The same general difference is shown for the two types of radiation by testing their comparative absorption by thin layers of paper, gold-leaf, silver-foil, and Dutch metal.

The following table is an example of the absorption of the radiation from a zinc plate and a thin layer of thorium oxide for thin tissue-paper:—

Thickness of layer of paper = ·0030 cm.
Potential-difference between plates = 50 volts.

	Current.	
No. of layers of paper.	Radiation from zinc.	Radiation from thorium oxide.
0	1	1
1	·57	·37
2	·35	·16
3	·20	·080
4	·12	·055

This method can also be used to compare the radiations from the various metals when made radio-active. In this way it was found that all the substances tried, viz., Cu, Pb, Pt, Al, Zn, brass, cardboard, paper, which had been made radio-active, gave out radiations of the same penetrating power. It was also found that the same type of radiation was given out from polished and dull surfaces, and that it was unaffected by the concentration of the radioactivity.

Since the same radiation is given out by all the metals and non-metallic substances like cardboard or paper, under varying conditions, we may conclude that either the substance itself which has been made radio-active plays no direct part in determining the kind of radiation, or that all exert exactly the same action.

The "excited" radiation is also of a more penetrating character than that given out by uranium, thorium, and the pitchblende derivatives radium and polonium.

Absorption of the Radiation in Air.

The absorption of the induced radiation in air was also determined. The method employed was similar to one previously used and described by the author * for determining the absorption of uranium radiation by different gases. A similar apparatus has been employed by Owens † for thorium radiation.

Two insulated parallel plates, kept a fixed distance apart, could be moved by means of a screw to different distances from the parallel radio-active surface. The radiation from the active surface passed through a circular opening in the lower plate, covered with thin aluminium-foil, and was stopped by the upper plate. The current between the two fixed plates for a large voltage was determined for different distances from the radio-active plate. If the radius of the active surface is large compared with the distance of the lower of the pair of plates from it, the current between the plates for a distance x of the lower plate from the active surface varies as $e^{-\lambda x}$, where λ is the coefficient of absorption of the radiation in the gas.

The following table gives the results obtained for the radiation from a lead surface which had been made strongly radio-active :—

* Phil. Mag. Jan. 1899, p. 124.
† Ibid. Oct. 1899, p. 378.

Lead Radiation.

Distance from surface.	Current.
d (=3 mms.)	1
d+6·25 mms.	·79
d+12·5 „	·59
d+18·7 „	·46
d+25 „	·35
d+31·2 „	·27
d+37·5 „	·21

The current is taken as unity when the measurements began at a distance $d=3$ mms. from the active lead plate.

For the purposes of comparison, the numbers obtained in a similar manner for thin layers of thorium oxide and uranium oxide on a bare plate are given below.

Thorium Radiation.

Distance.	Current.
d (=2·25 mms.)	1
d+5 mms.	·73
d+10 „	·50
d+15 „	·35
d+20 „	·25

Uranium Radiation.

Distance.	Current.
d (=2·25 mms.)	1
d+ 2·5 mms.	·685
d+ 5 „	·445
d+ 7·5 „	·296
d+10 „	·188
d+15 „	·088
d+20 „	·059

The curves in fig. 6 show the results graphically. It will be seen that the intensity of the radiation falls off approximately in a geometrical progression as the distance increases in arithmetical progression. Curves of absorption of thorium radiation in air at different pressures have been obtained by Owens *.

The distances through which the three types of radiation from uranium, thorium, and active lead pass through air at ordinary pressures and temperatures before the intensity is reduced to one-half its value, are about 4, 10, and 16·5 mms. respectively.

* Owens, Phil. Mag. Oct. 1899.

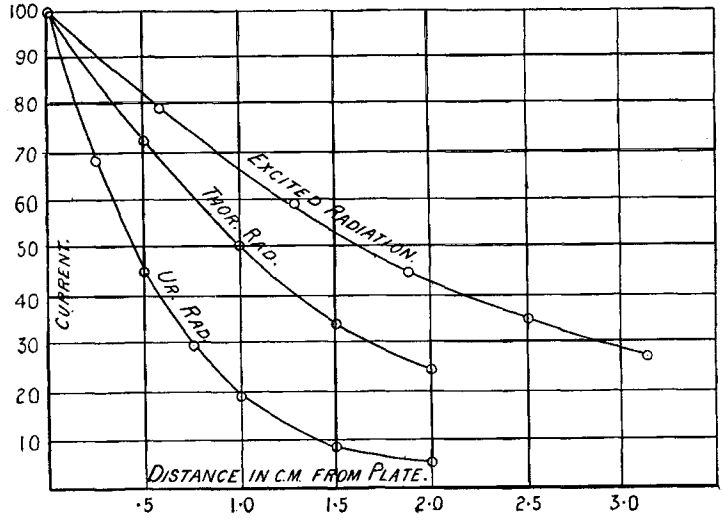
Assuming that the intensity falls off as $e^{-\lambda x}$, the values of λ for the types of radiation are given below.

	Value of λ.
"Excited" radiation	·42
Uranium „	1·6
Thorium „	·69

The order of absorption in air of the above three types of radiation is the same as for aluminium and paper.

The "excited" radiation is of a more penetrating kind than the easily absorbed type (the α radiation) * given out by uranium, but much less than the β type. The radiations from radium and polonium are also more readily absorbed in air than the excited radiation is.

Fig. 6.



Duration of the Radioactivity.

If a plate or wire which has been made radio-active is removed from the action of the thorium, the intensity of the radiation diminishes according to a very simple law.

A large number of experiments have been made on the duration of the induced radioactivity in various substances under varying conditions. A typical table of the results obtained is given below for a rod of brass which has been

* Rutherford, Phil. Mag. Jan. 1899, p. 116.

made active. In order to test the rate of decay of the intensity, the active rod was placed inside a cylinder and concentric with it. The current between the two cylinders for a potential-difference of 50 volts was measured in the usual manner, and at intervals of several hours.

Length of rod = 31·5 cms.
Diameter = ·40 cms.
Testing-cylinder, inside diameter = 7·3 cms.

Time in hours.	Current.
0	1
7·9	·640
11·8	·474
23·4	·196
29·2	·138
32·6	·103
49·2	·0370
62·1	·0186
71·4	·0086

The value of the maximum current, which is taken as unity, was $1{\cdot}6 \times 10^{-11}$ amperes.

Fig. 7 shows graphically the results obtained. The results show that the current through the gas (which is proportional to the intensity of the radiation) diminishes in geometrical progression with the time. The time taken for the intensity of the radiation to fall to half its value is about eleven hours. If I_0 be the intensity at the beginning, the intensity I after a time t is given by

$$I = I_0 e^{-Lt},$$

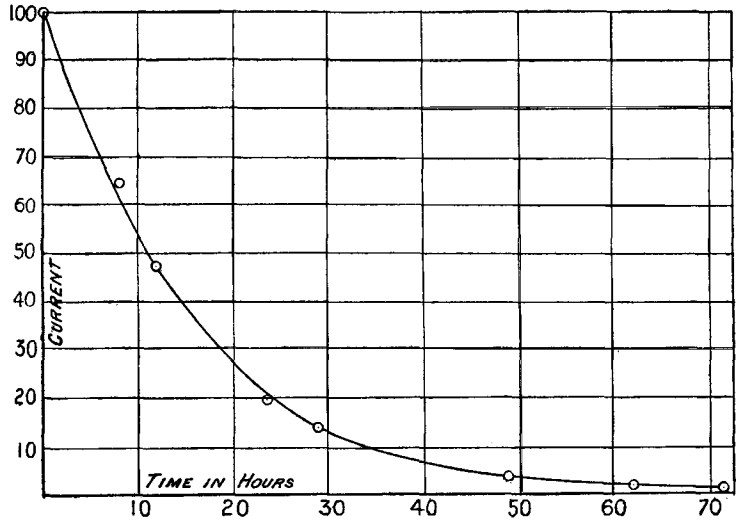
where L is a constant.

The above law appears to hold accurately for all substances made radio-active. No difference in the rate of decay has been observed, whether the radiation is on a plate of large area or concentrated on a fine wire. The rate of decay is also independent of the substance made radio-active. A piece of paper, mica, or metal, all give the same rate of loss of intensity. As far as experiments have gone, the rate of decay is unaffected by the pressure of the gas surrounding it, or whether the air is dry or full of moisture. The same rate of decay has always been obtained under all the conditions tried, provided the surface is not acted on mechanically or by chemicals.

The mean value of L deduced from the above results is

$$L = {\cdot}0000189.$$

Fig. 7.



In a previous paper * I have shown that the radio-active "emanation" from thorium compounds quickly loses its radio-active power.

The intensity in that case falls to half its value in about one minute, while the intensity of the "excited" radiation falls to half its value in about eleven hours, or one decays 660 times faster than the other. The law of falling off of intensity is the same in the two cases.

On page 166 it has been shown that the current i (for a "saturating" E.M.F.) between two cylinders is given by

$$i=\frac{SI_0}{A}\{1-e^{-\lambda(b-a)}\},$$

with the same notation as before.

The intensity I of the radiation after a time t is given by

$$I=I_0 e^{-Lt},$$

and the total quantity of electricity passing between the cylinders during the time taken for the intensity to fall to zero is given by

$$Q=\int_0^\infty i\,dt=\frac{SI_0}{A}\{1-e^{-\lambda(b-a)}\}\int_0^\infty e^{-Lt}dt$$

$$=\frac{SI_0}{LA}\{1-e^{-\lambda(b-a)}\};$$

* Phil. Mag. Jan. 1900.

if i_0 = initial current, it is clear that

$$Q = \frac{i_0}{L}.$$

In the case given in the last table, the initial current was $1{\cdot}6 \times 10^{-11}$ amperes, and the value of L = ·0000189; therefore the total quantity of electricity passing between the cylinders is equal to $8{\cdot}5 \times 10^{-7}$ coulombs.

The total quantity of electricity separated, if the radiation has been completely absorbed in the gas, is obviously

$$\frac{1}{1-e^{-\lambda(b-a)}}$$

of this quantity.

In the above case, a = ·20 cm., b = 3·65 cms., λ = ·42.

Therefore quantity passing between cylinders = $11{\cdot}1 \times 10^{-7}$ coulombs.

Increase of Induced Radioactivity with Time.

If a plate or wire is exposed to the action of thorium oxide in a closed vessel, the radioactivity at first increases nearly proportionally with the time, and then more slowly, finally tending to a maximum value after several days' exposure.

The table given below is an example of the results obtained for a square zinc plate, area 86 sq. cms., exposed in a metal vessel, with a potential-difference of 300 volts between thorium and surface to be made active. The plate was removed from the action of the thorium at intervals for sufficient time to determine the current produced by it between two charged parallel plates, as in fig. 1.

Time of exposure in hours.	Current.
1·58	·063
3·25	·105
5·83	·289
9·83	·398
14·00	·586
23·41	·773
29·83	·834
47·00	·898
72·50	·951
96·00	1·00

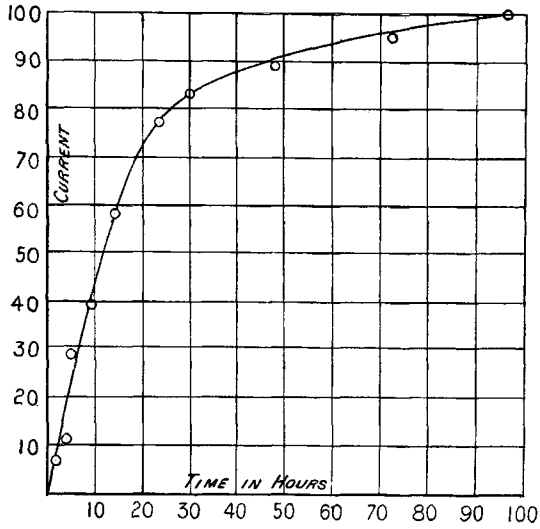
The current after four days' exposure is taken as unity, as the rate of leak had nearly reached its maximum value.

The maximum value of the current produced by the active

plate between two test-plates 4 cms. apart was $1{\cdot}7 \times 10^{-11}$ amperes.

Fig. 8 shows the results graphically. From the table it will be seen that the intensity has reached half its final value in about twelve hours.

Fig. 8.



We will now consider the conditions which influence the increase of the intensity of radiation from a given surface exposed to the action of a thorium compound. We will suppose that the surface to be made radio-active is negatively charged.

Two opposing actions are evidently at work. Fresh radio-active particles are being continually carried to the plate, while the intensity of the radiation given out by the active surface continually diminishes, owing to the radiation of energy. A steady state will be reached when the rate of increase of intensity due to the supply of fresh radio-active particles is equal to the rate of decrease of the intensity due to the radiation of energy from the active surface.

Let I be the intensity of the radiation at the surface of the plate at any time. The rate of diminution of the intensity is equal to λI, since the intensity I at any time is given by

$$I = I_0 e^{-\lambda t}$$

and

$$\frac{dI}{dt} = -\lambda I.$$

Let q be the rate of increase of the intensity due to the steady supply of radio-active material. Then

$$\frac{dI}{dt} = q - LI$$

or

$$\log_e(q - LI) = -Lt + A.$$

But $I = 0$ when $t = 0$.

Therefore

$$A = -\frac{1}{L}\log_e q.$$

Therefore

$$\log_e \frac{q - LI}{q} = -Lt$$

or

$$I = \frac{q}{L}(1 - e^{-Lt}).$$

When Lt is very large, the maximum value of the intensity I_0 is given by

$$I_0 = \frac{q}{L} \quad \text{and} \quad \frac{I}{I_0} = 1 - e^{-Lt};$$

or the equation representing the rise of intensity of the radiation is the same as the rise of an electric current in a circuit of constant self-induction.

The curve which is shown in fig. 8 is in rough agreement with this equation. For example, the intensity of the radiation has risen to half its value in about twelve hours. Now $e^{-Lt} = \frac{1}{2}$ when $t = 11$ hours, *i. e.*, according to theory, the current should have reached half its value in about eleven hours.

There is a divergence between the theoretical and observed results in the first part of the curve. The rate of increase of intensity is slower at first than the theory would suggest. It is probable, however, that the rate of supply of radio-active material does not reach a steady value for a considerable time after the exposure of the plate, and such a cause would account for the results observed. Other results obtained, under different conditions, all show too small a value of the intensity for the first few hours of exposure.

We have so far assumed that the radio-active particles were conveyed to the surface under the influence of an electric field. The equations which have been given will, however, apply equally well to the case of diffusion. If no electromotive forces are acting, the radio-active particles

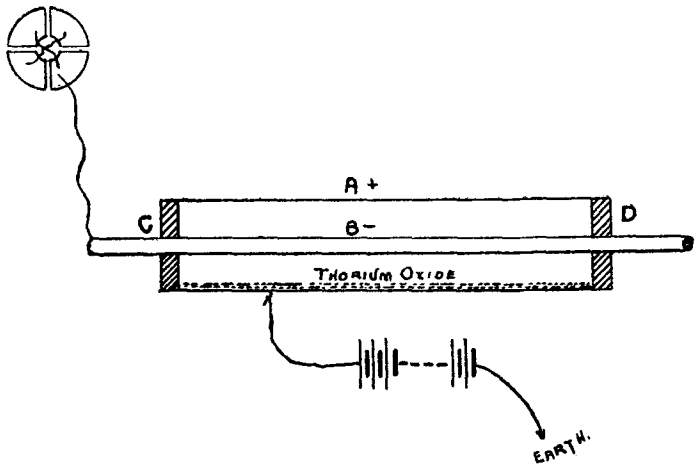
diffuse through the gas and adhere to the surface on which they impinge. A steady state will be reached when the rate of supply of fresh radio-active particles due to diffusion is balanced by the decay of the radiation from the surface. The maximum intensity of the radiation on any surface in the neighbourhood of a thorium compound is thus proportional to the number of radio-active particles that reach it by the processes of diffusion.

Effect of E.M.F. on the Amount of Radioactivity.

The amount of induced radioactivity in a given time increases with the voltage for small voltages, but soon reaches a point beyond which large increases in the E.M.F. have a very small effect on it. In order to investigate the relation in detail, the following arrangement was employed :—

Two insulated concentric brass cylinders A and B (fig. 9) were used, of diameters 5·5 and ·7 centim. respectively. The

Fig. 9.



ends were closed with paraffin stoppers C and D. The cylinder A was connected with the + pole of a battery, the other pole of which was to earth. The cylinder B was connected to the electrometer in the usual manner. A layer of thorium oxide in a paper envelope was placed along the bottom of the cylinder A. The whole was exposed to the action of thorium oxide for three days. The intensity of the radiation given out by B had, after that interval, nearly reached its maximum value.

The following measurements were made for each experiment at different voltages :—

(1) The current between A and B was measured with the thorium oxide in the cylinder.

(2) After the thorium oxide had been removed and the air blown out, the current between A and B was again determined.

(3) The cylinder B was then removed and a non-active one of the same dimensions substituted and the current again observed.

The electrometer was brought to the same sensitiveness every day by means of a Thomson Replenisher. For rapid rates of discharge, a condenser of ·001 microfarad capacity was placed in the electrometer circuit.

The current (3) includes the small leak (if any) over the insulators plus the current due to the radioactivity produced in the end paraffin stoppers. The current (2) was due to the radio-active cylinder B, together with the current (3). The current (1) was due to the "emanation" from thorium oxide plus (2) and (3).

From these three observations it was therefore possible to determine :—

(*a*) The rate of discharge due to the thorium alone ;

(*b*) The rate of discharge due to radio-active cylinder B alone ;

(*c*) The rate of discharge due to radioactivity on the sides and ends of vessel.

In the following table the results are given for different voltages between cylinders.

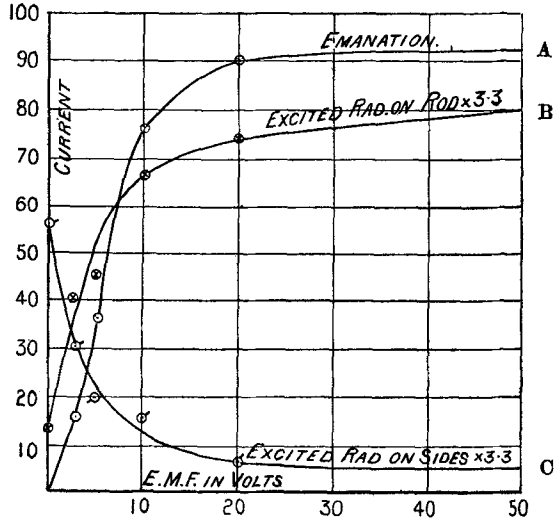
Results are in divisions per second of electrometer-scale.

Voltage.	Emanation.	Radioactivity. on cylinder B.	Radioactivity on sides and ends.	Total Radioactivity.
0	.....	1·32	5·58	6·90
3	5·3	4·02	3·06	7·08
5	12·2	4·53	1·97	6·50
10	25·3	6·69	1·51	8·20
20	30·0	7·40	·59	7·99
310	32·2	9·91	·75	10·66

Fig. 10 shows the results graphically. Curve A shows the variation of the current due to the "emanation" with voltage ;

curve B, the variation of the amount of induced radioactivity on inside cylinder; and curve C, the variation of the amount of radioactivity on the sides and ends. The ordinates of curves B and C are increased three times in order to show them on about the same scale as A. It will be observed that the shapes of the curves A and B are similar. The "knee" of both curves occurs for about the same voltage. The

Fig. 10.



curve C shows that as the voltage diminishes, the amount of radioactivity on the sides and ends increases, reaching a maximum when the voltage is zero. The currents due to the radioactivity of B, given in the third column of the table, are for 50 volts between the cylinders. The value given for 310 volts is probably too large, as it was measured for 310 volts between plates, instead of 50 as in the other experiments.

In the fifth column is given the total current due to the radioactivity on ends and sides plus the action of cylinder B. It will be observed that the resulting values are not very different, except the value for 310 volts, which, for reasons above explained, is probably too large.

It looks as if a certain number of radio-active particles were given out from the thorium and that these were carried to various parts of the vessel, the effect due to the whole number being about the same as if they were all concentrated on the negative electrode.

Case of Diffusion of Radio-active Particles.

The case where no voltage is acting is one of special interest, for there the diffusion of the radio-active particles is alone operative. A loose layer of paper was placed over the paper envelope containing the thorium oxide. The paper envelope bent into the arc of a circle covered about one quarter of the circumference of the cylinder. The following numbers give the rate of leak, in divisions per second, due to the radioactivity on different portions of the vessel :—

Radioactivity on inside cylinder ...		1·32	divisions per sec.
„	on paper	2·26	„
„	on outside cylinder and stoppers	3·32	„
Total radioactivity		6·90	„

The current due to the total radioactivity is thus about the same as the current when 20 volts acts between cylinders. The experiments on the effect of voltage extended for more than a month, and some of the results showed that the thorium oxide was not a constant source of radiation during the whole of that time. The variations were not, however, sufficiently large to obscure the general nature of the results.

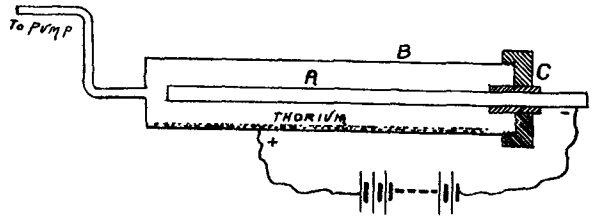
Effect of Pressure on Radioactivity.

The diminution of the pressure of the gas from 760 to 20 millim. had very little action on the amount of "excited" radioactivity on the − charged electrode.

The following apparatus was employed :—

A brass cylinder B (fig. 11 *a*), with an ebonite stopper C,

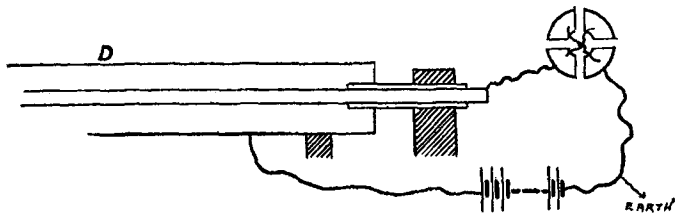
Fig. 11 *a*.—Exciting Apparatus.



through which passed a brass rod A, was connected with a mercury pump. The thorium oxide inside a paper envelope was placed inside the cylinder. B was connected to + pole of a battery of 50 volts, and C to − pole. The apparatus was

exhausted to the required pressure as rapidly as possible, and rod A exposed for several hours. The rod was then removed, and the current due to its radioactivity tested inside another cylinder D (fig. 11 *b*). The battery and electrometer

Fig. 11 *b*.—Testing Apparatus.



connexions are seen in figure. On account of the press of other work, it was not found possible to take observations at regular intervals, but the table given below suffices to show the general nature of the results.

Pressure.	Time of Exposure.	Divisions per second.	Divisions per sec. Interpolated values.
millim.	hours.		
760	5·25	2·37	13·1
175	20	9·83	13·9
16	4·1	2·15	15·1
4·5	5·4	1·96	10·3
1·7	4·8	·72	4·5
·45	14·3	·38	·65
·04	25	·34	·44

The third column gives the current in divisions per second due to radio-active rod in the testing vessel. In the fourth column are given the divisions per second corresponding to an exposure of the rod for the same time (three days) in each case, at the particular pressure. The results are interpolated from the first two columns with the help of the curve given in fig. 8. The results must only be considered approximate, and merely serve to give a comparative estimate of the radioactivity at each pressure. The general results are clear. The radio-activity is about the same at 16 millim. as at 760 millim. Between the pressures of 16 and 4·5 millim. the amount begins to diminish, until at ·45 millim. it is only $\frac{1}{20}$ of the

value at atmospheric pressure. Still further diminution of pressure does not have much effect.

A special experiment on the distribution of the radio-activity at low pressures throws some light on the phenomena. If we expose a rod charged — at a low pressure to the action of thorium, it will be found that the rod is only slightly radio-active, while the top of the paper over the thorium oxide and the sides of the vessel are strongly radio-active. At atmospheric pressure, other conditions remaining the same, it will be found that most of the radioactivity is confined to the rod, and only a slight amount is produced on the paper and sides of the vessel. It appears as if the radio-active particles are unable to be all carried to the negative electrode at low pressures. This may be due to the increased rate of diffusion of the active particles at low pressures, or more probably to the small number of ions produced by the "emanation" at low pressures.

It is found that the current through the gas due to the "emanation" falls off nearly proportionally to the pressure, so that the number of ions present between cylinders at low pressures is a very small fraction of those at atmospheric pressure.

The following table gives results of the variation of the current, due to the emanation, with pressure of air in the apparatus of fig. 11 *a*.

Pressure of Gas. millim.	Current due to Emanation.
760	1
587	·819
402	·582
214	·297
145	·203
93	·133
25	·046

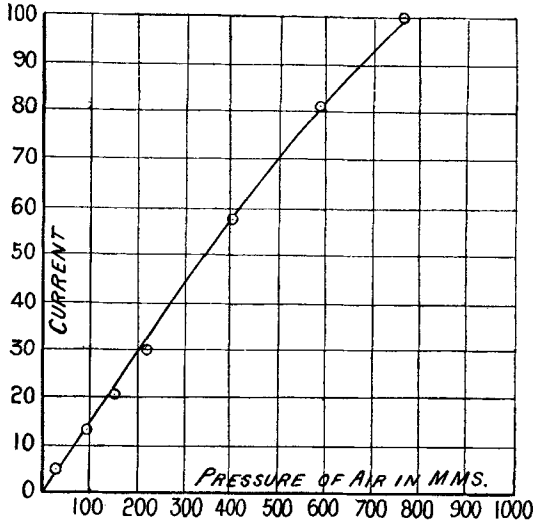
Fig. 12 shows the results graphically. The curve is nearly a straight line. If the conveyance of the radio-active particles to the electrode is due to the movements of the ions between cylinders, at low pressures the number of ions may be too small to be effective in that respect.

Effect of Gases.

The apparatus shown in fig. 11 was used. The amount of radioactivity produced on the central rod was not very different whether the gas was hydrogen, air, or carbonic acid.

No definite difference was observed whether the gas was free from water-vapour or not. The amount of current due to the emanation from thorium oxide was found, however, to vary

Fig. 12.



greatly with the gas. Taking the current due to air as unity with 50 volts acting, the currents due to the "emanation" were

Air	1
H	·35
CO_2	1·1

These numbers are not necessarily proportional to the ionization constants of the gas, as the current produced depends on the relative absorption of the rays between the cylinders.

These results, together with those obtained for lowering of the pressure in air, show that there is no evident quantitative connexion between the current due to the emanation and the amount of induced radioactivity.

Chemical and Mechanical Actions on the Radio-active Surface.

We have previously considered the conditions which govern the production and decay of the induced radioactivity. We will now describe some experiments that have been made to

try and throw some light on the question as to what the induced radioactivity is really due to.

If the radioactivity is caused by some radio-active dust deposited on the substance, we should expect to find evidence of it by examining the surface with a microscope, or by noting whether there is any increase in weight. A fine piece of platinum wire, which had been carefully weighed, was made strongly radio-active by five days' exposure to thorium oxide covered over with paper. Within the limits of accuracy of the balance no certain variation of the weight could be detected. The increase of weight, if any, was certainly less than $\frac{1}{20}$ of a milligramme. On examination by a microscope no collection of dust particles on the surface could be observed. We may conclude from this experiment, that if the radioactivity is due to the deposition of radio-active particles on the surface, these particles must be extraordinarily radio-active compared with their weight. A rough estimate shows that the radioactivity of the surface-layer must be at least a million times greater than that of uranium or thorium.

The amount of radiation from an active surface is always lessened by mechanical actions, such as rubbing the surface with a cloth or fine sand-paper. In order to completely remove the radioactivity, it is necessary to remove the surface-layer by long scouring with sand- or emery-paper.

A blast of air directed against a radio-active plate has no appreciable effect on the amount of radiation given out.

A radio-active platinum wire or plate can be heated white-hot without much altering the amount of radiation given out from it. A strongly active fine wire is more affected than a plate; but that is probably chiefly due to action of the flame-gases upon it.

Chemical Actions.

The radioactivity of a platinum plate is not much affected by dipping it in water, caustic soda, or nitric acid, whether hot or cold. Sulphuric or hydrochloric acid has the power of rapidly destroying the intensity of the radiation in a few minutes. A copper-sulphate solution, if only slightly acid, does not act on the wire rapidly. The following example of a test shows the effect of several of the solutions on a radio-active platinum plate. After each immersion the plate was washed in water and dried over a Bunsen-flame. After exposure of 4 minutes to gradually heated water and 2 minutes to boiling water, the rate of discharge fell from 100 divisions

in 15·5 secs. to 100 divisions in 20 secs. After 5 minutes' boiling in caustic soda, the rate of discharge fell to 100 in 27 secs. After 10 minutes' exposure to strong hot nitric acid, the rate of discharge was cut down to one-half its previous value. Dilute sulphuric acid reduced the rate of discharge to one-half in 10 secs. and one-quarter in 60 secs. Both hydrochloric and sulphuric acids are more powerful in destroying the radio-active power than the other solutions examined. In the case of water and caustic soda, the small diminution of intensity appears to be due as much to the mechanical action of the bubbling as to the chemical action on the surface.

The question now arises, whether the loss of radioactivity of the active plate by immersion in solutions is due to the destruction of the radio-active power of the particles or their removal from the plate to the solution. A fine platinum wire, very strongly active, was placed in a few drops of dilute sulphuric acid for several minutes. The wire lost a large proportion of its radioactivity. The dilute acid was then evaporated down to dryness in a sand-bath, and on examination it was found that the residue on the glass surface was strongly active. We may conclude from this experiment that the radioactivity of the particles is not destroyed, but that they pass into solution, and that on evaporating the solvent the substance still remains.

Some experiments were tried to see whether a plate preserved its radio-active power when a layer of copper was electrolytically deposited upon it. A radio-active platinum wire was made a cathode in a copper-sulphate solution, and a current of about half an ampere passed through for 1 minute. The radioactivity was diminished to about ·7 of its value when tested in the usual way. After washing the wire in water, it was allowed to stand some time in air, and the rate of diminution of the radioactivity observed. The intensity diminished more rapidly at first than for an unacted-on wire; but after 10 hours the rate of diminution became normal. The more rapid decrease at first is probably due to the dilute sulphuric acid which remained in the pores of the copper deposit. When the platinum wire was made the anode in a copper-sulphate solution, the radioactivity rapidly diminished. The action in this case was probably due to the production of sulphuric acid at the surface of the anode by the passage of the current which dissolved the radio-active material on the platinum plate.

Discussion of the Results.

Before entering on the question of the cause and nature of induced radioactivity, a brief review may be given of the results obtained :—

(1) All thorium compounds examined produce radioactivity in substances in their neighbourhood, if the bodies are all uncharged. With charged conductors the radioactivity is produced on the — charged body. In strong electric fields, the radioactivity can be concentrated on the surface of thin wires. Thorium oxide is the most active of the thorium compounds in causing radioactivity, but loses its power if it is heated for several hours at a high temperature.

(2) The power of producing radioactivity is closely connected with the presence of the "emanation" from thorium compounds, and is in some way dependent upon it.

(3) The radiation excited in bodies is homogeneous, and of a more penetrating character than the radiations from thorium or uranium. The radiation is confined to the surface of the substance, and is independent of whether the substance is a conductor or non-conductor and of the nature of its surface.

(4) The intensity of the radiation emitted falls off in a geometrical progression with the time, decreasing to half its value in about 11 hours. The decay of intensity is independent of the state of concentration of the radioactivity or the nature of the substance.

(5) The amount of induced radioactivity increases at first nearly proportional to the time of exposure, but soon tends to a value when the intensity of the radiation varies very little with increase of the time of exposure.

(6) The amount of induced radioactivity produced in a given time on a conductor depends on the potential-difference between the electrodes, and tends to a constant value for large E.M.F.'s.

(7) The amount of radioactivity is independent of the pressure of the gas, except at low pressures when the amount on the — charged conductor decreases with the pressure. The amount is not much affected whether the gas is hydrogen, air, or carbonic acid.

(8) No increase of weight has been observed by making a body radio-active. The radiation from a platinum wire is not much altered by placing the wire in a flame, hot or cold water, or nitric acid. Hydrochloric and sulphuric acids rapidly remove the radioactivity from its surface. The solution, when evaporated, leaves the active portion behind.

Three possible explanations of the phenomena of induced

radioactivity naturally present themselves:—(*a*) That the radioactivity is due to a kind of phosphorescence excited in the substance by the radiation from thorium; (*b*) or to the deposition of the + gaseous ions produced in the gas by the "emanation"; (*c*) or to the deposition of particles of a radio-active material emitted by thorium compounds.

The hypothesis that the radiation is a kind of phosphorescence will not explain the results observed, since substances are made radio-active outside the incidence of the radiation, and the radioactivity can be concentrated on the — electrode. The question as to whether the induced radioactivity is due to the deposition of a foreign substance on bodies, or to the action of the + ions produced in the gas, or a combination of both, is difficult to decide with certainty from the experimental evidence. The theory that the + ions produced by the emanation are responsible for the radioactivity, at first sight seems to explain many of the results. Since the radio-active particles of the emanation are very small, the intensity of the radiation must be very great near them; and in consequence of this, ions may not only be produced, but the charges on the ions set in violent vibration: these + ions would be carried to the negative electrode, and gradually dissipate the energy of their vibration by radiation into space. On this theory, however, it is difficult to explain the variation of radioactivity with pressure. At low pressures, the experiments show that the total radioactivity produced is much the same as at atmospheric pressure, but the — electrode receives only a small proportion of the radio-active particles On the theory that the radio-active particles are + ions, we should expect them in a strong field to be all carried to the — electrode. Another experiment on the variation of the amount of radioactivity with distance also does not fall in readily with this view. The amount of radioactivity was found to be practically the same whether the distance from the radio-active surface was 3 mms. or 3 cms. In the latter case, the number of + ions produced by the emanation is much greater than in the former; but the amount of radioactivity is unaffected.

The theory that the radioactivity is due to a deposition of radio-active particles from the thorium compounds affords a general explanation of all the results; but the difficulty is to advance a satisfactory reason for the particles obtaining the + charge which they must possess in order to be moved to the — electrode in an electric field. If we suppose the radio-active particles from thorium compounds emitted at a uniform rate, independent of the nature and pressure of the gas, we should

expect to obtain the same total amount of radioactivity spread over a vessel due to diffusion of the particles, as can be obtained by concentration of all the radio-active particles on the — electrode; and the amount should be independent of the pressure and nature of the gas, provided it does not act on the thorium. Some experiments seem to point to the conclusion that the radio-active particles are not charged till they diffuse out into the gas, but that they gain a + charge in the course of time. A possible explanation is that the + charge is obtained by the diffusion of the ions to the surface of the particles. Since there is reason to believe that the — ions in most cases move faster than the + ions in an electric field, there is always an excess of + ions in the gas, and the particles in the gas thus tend to become positively charged. On this supposition, the diminution of the amount of radio-activity on the — electrode at low pressures is due to the fact that there is not a sufficient number of ions in the gas to charge the particles, which thus diffuse to the sides of the vessel.

As far as experiments have gone, the power of exciting radioactivity appears to be confined to thorium compounds. Neither uranium nor radium nor polonium has so far shown any trace of action; but the specimens * of radium and polonium used were not very radio-active and contained considerable amounts of impurity. A plate made radio-active is not able to excite any appreciable radioactivity in another plate near it. I have tested the + and — electrodes after the passage for several hours of a strong current between them due to Röntgen rays, flames, and discharge from points, but no trace of radioactivity on them has been observed.

Macdonald Physics Building,
McGill University, Montreal,
Nov. 22nd, 1899.

* As this paper was passing through the press the *Comptes Rendus* of Nov. 6th was received, which contains a paper by Curie and a note by Becquerel on the radiation excited in bodies by radium and polonium. Curie has used specimens of these substances 10,000 to 50,000 times more radio-active than uranium and the phenomena observed are, in some respects, similar to those exhibited by thorium compounds; but there are not sufficient data on which to base any comparison. No mention is made of the effect of an electric field, or whether there is an "emanation" from radium and polonium, as there is from thorium compounds. Curie concludes that the results obtained are due to a kind of phosphorescence excited by the radiation; while in the case of thorium the author has shown that such a theory is inadmissible. Further experiments on the comparison of the radioactivity produced by thorium with that produced by radium and polonium will be of interest.