

proportions in which the radiation of longer period and shorter period are present in the total radiation depends on the radiating surface, other things being the same. In the case of the sooted wire, the quantity of long-period radiation is, in proportion, far in excess of that proceeding from a bright metallic polished surface. Consequently, with the *same* total electric energy supplied to both wires, the glass tube containing the sooted wire becomes *very much* hotter than the tube containing the bright wire.

It has also been pointed out that with a substance like glass, conducting badly and somewhat diathermanous, it is impossible to tell how much heat is returned to the radiating wire from the interior skin of the tube, which no doubt rises to a high temperature during the experiment. To a certain extent, therefore, the results which we have recently obtained must be considered as not strictly comparable with those formerly obtained, in which a metallic envelope cooled with water was used.

The absolute value of the radiation observed ought to be somewhat lower in amount than would have been found had the enclosing envelope been of metal properly kept cool, and the disturbance from this cause must have been relatively greater in the case of the dull than in the case of the bright wire.

I cannot conclude without expressing my warmest thanks to my assistant, Mr. Evans, whose aid has been invaluable. Without the help which he has given me the experiments could not have been carried out at all. The investigation is still in progress, indeed, the present paper must be regarded as a description of preliminary trials; I hope before long, however, to obtain further results in the same direction of considerable interest. The temperatures shown on the curves and in the tables have been obtained partly by direct experiment and partly by using the results of other experimenters who have given comparisons between platinum resistances and temperatures. They must only be regarded as approximations, and may possibly require to be corrected. The amount of the correction cannot, however, alter the general conclusion that the polished surface is much more economical for the production of light than the sooted surface; and that as far as our experiments have gone, the polished platinum surface and the sooted surface are practically at the same temperature when they present a similar appearance to the eye.

LXIV. *The Cause and Nature of Radioactivity.*—Part II.
By E. RUTHERFORD, M.A., D.Sc., *Macdonald Professor of Physics*, and F. SODDY, B.A. (Oxon.), *Demonstrator in Chemistry, McGill University, Montreal* *.

CONTENTS.

- I. Introduction.
- II. Method of Measuring Emanating Power.
- III. The De-emanation of Thoria and the Regeneration of the Emanating Power.
- IV. The Effect of Conditions upon Emanating Power.
- V. The Cause of the Emanating Power of Thorium.
- VI. The Chemical Nature of the Emanation.
- VII. The Nature of Emanating Power.
- VIII. The Excited Radioactivity from Thorium.
- IX. Further Theoretical Considerations.

I. *Introduction.*

THE investigation of the radioactivity of thorium, detailed in the first part of this communication†, arose out of an examination of the power possessed by thorium compounds of giving out a radioactive emanation. The nature of this property and its relation to the radioactivity of thorium remain to be considered.

A short résumé of what was known at the commencement of the work may be of interest. Thorium radioactivity was discovered by Schmidt and Curie independently in 1898, and Owens in the following year investigated its nature in detail (*Phil. Mag.* 1899, p. 360). He observed the inconstancy of the radiation and the effect of air currents in reducing its value. The discovery of the thorium emanation which explained these results, and its power of exciting activity on surrounding matter, followed shortly after (Rutherford, *Phil. Mag.* 1900, pp. 1 & 161).

It was shown that the radiation from the emanation decays rapidly, but at a perfectly defined rate, falling to about one-half the original value at the end of one minute. The emanation passes unchanged through cotton-wool, weak and strong sulphuric acid, and aluminium and other metals in the form of foil, but not through an extremely thin sheet of mica. The emanating power of thoria is independent of the surrounding atmosphere, but is destroyed to a large extent by intense ignition, and does not return when the substance is kept.

There is a very close connexion between the excited radioactivity produced by thorium compounds and the emanation.

* Communicated by the Authors.

† For Part I. see *Phil. Mag.* Sept. 1902.

It was shown that the amount of the former produced under various conditions was proportional to the amount of the latter, and if the emanating power of thoria be destroyed by ignition, its power to excite radioactivity correspondingly disappears. Simultaneously with the appearance of the papers referred to, Curie showed that radium also possessed the power of exciting activity on surrounding objects. Later, Dorn (*Abk. der Naturforsch. Ges. für Halle-a.-S.*, 1900) repeated the work quoted for thoria, and extended it to include two preparations of radioactive barium compounds (radium) prepared by P. de Haen, and a preparation of radioactive bismuth (polonium). He found that radium, but not polonium, gave an emanation, especially on warming, and this possessed the power of exciting activity on surrounding objects. Radium and thorium are in this respect completely analogous and different from other radioactive substances, but the phenomena in the two cases are quite different. The emanation from radium retains its activity for many weeks, while the excited radioactivity it produces, on the other hand, decays much more rapidly than that from thorium.

One of the most interesting advances in this connexion was made during the progress of the work by Elster and Geitel (*Phys. Zeit.* 1901, ii. p. 590), who found that it is possible to produce excited radioactivity from the atmosphere, without further agency, by simply exposing a wire highly charged to a negative potential in the air for many hours, and that this also possesses the property of being dissolved off by acids, and of being left behind unchanged on the evaporation of the latter. But here again the rate of decay is different from that of the excited radioactivity produced by thorium.

At the commencement of the work the presumption seemed to be in favour of considering emanating power as a separate phenomenon not directly connected with the ordinary radioactivity of thorium. The former could be destroyed in thorium oxide by ignition without reducing the latter. Later many external conditions were found to affect the value of emanating power without influencing the radioactivity. The nature of the phenomenon had been fully examined from this point of view with very puzzling results, but the conclusion was arrived at that emanating power is probably the manifestation of a change of the nature of a chemical reaction.

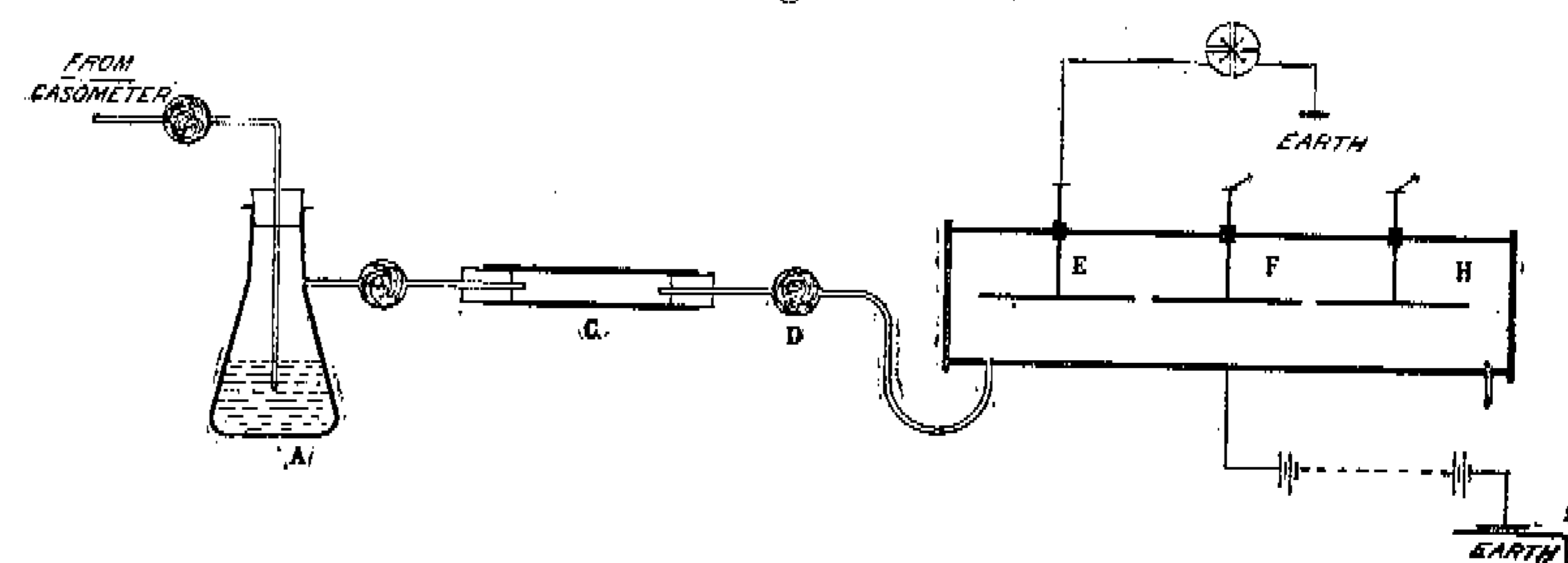
The discovery of ThX and its continuous production, however, revealed the true interpretation of the results, and enables a fairly complete explanation of the phenomenon to be given.

II. Method of Measuring Emanating Power.

The emanation from thorium (and from radium) behaves in all respects like a temporarily radioactive gas, and diffuses rapidly through porous substances, as, for example, thick cardboard, which are completely opaque to the straight line radiation. Each particle of the emanation behaves as if it were a radiating centre, producing charged carriers throughout the gas in its neighbourhood. The emanation passes through plugs of cotton-wool and can be bubbled through liquids without appreciable loss of radioactivity, whereas the charged carriers, produced by the emanation in common with the straight line radiation from radioactive substances, on the contrary, completely disappear on passing through a plug of cotton- or glass-wool, or by bubbling through liquids. The means of eliminating the effects of the straight line radiation and of measuring the amount of the emanation alone thus suggest themselves. Air passed over uranium or other non-emanating radioactive substance will no longer conduct a current after passage through cotton-wool. The conductivity in the case of thorium, however, will persist, and afford a measure of the amount of emanation present.

Fig. 1 shows the experimental arrangement for comparing

Fig. 1.



the emanating power of substances. These are placed in the form of fine powder in a shallow lead vessel inside the glass cylinder, C, 17 cm. in length and 3.25 cm. in diameter, provided with indiarubber corks. A current of air from a large gas-bag, after passing through a tube containing cotton-wool to remove dust particles, bubbled through sulphuric acid in the vessel A. It then passed through a bulb containing tightly packed cotton-wool to prevent any spray being carried over. The emanation mixed with air was carried from the vessel C through a plug of cotton-wool, D, which completely removed all the charged carriers carried with the emanation. The latter then passed into a long brass cylinder 75 cm. in

length and 6 cm. in diameter. The cylinder insulated on paraffin blocks was connected to one pole of a battery of small lead accumulators, the other pole of which was connected to earth. Three electrodes E, F, H, of equal length were placed along the axis of the cylinder. The current through the gas was measured by means of a Kelvin electrometer of the White pattern. The electrometer and connexions were suitably screened by means of wire gauze connected to earth. An insulating key was arranged so that either of the electrodes E, F, H, or all of them together, could be rapidly connected to one pair of quadrants of the electrometer, the other two being always connected to earth. The measurements were carried out in the usual way by observing the rate of movement of the electrometer-needle after the one pair of quadrants were connected with the electrodes. On placing the emanating substance in C and continuing the air current for several minutes at a constant rate, the current through the gas due to the emanation attains a steady state. The number of divisions of the scale passed over per second may be taken as a measure of the current.

With this apparatus the emanation from 10 grams of ordinary thorium oxide produces a current of 3.3×10^{-11} amperes between the three electrodes connected together and the cylinder. With the electrometer working at an average sensitiveness, this corresponded to a deflexion of 100 divisions of the scale in 12 seconds, so that one-hundredth part of this current could be readily measured—that is, the emanation produced by one-tenth of a gram of thorium oxide. The electrometer one hundred times more sensitive than this failed to detect the presence of an emanation or radioactivity in the oxides of tin, zirconium, and titanium, the other elements of the same group in the periodic table.

Rate of Decay of the Radiation from the Emanation.—The three electrodes E, F, H, were used to compare the “rates of decay” of the radiations from the emanations of different substances. In the previous papers quoted, it has been shown that the radiating power of the thoria emanation falls to half its value in about a minute. In consequence of this, the current observed for the electrode E is greater than for the electrode H. Knowing the velocity of the current of air along the cylinder and the respective currents to the electrodes E, F, H, the rate of decay of the radiation can be readily deduced. If, however, we merely require to compare the rate of decay of one emanation with another, it is only necessary to compare the ratio of the currents to the elec-

trodes E, F, H in each case, keeping the current of air constant. If the ratio of the currents is the same we may conclude that the radiating power of each diminishes at the same rate. The comparison of the emanation is thus rendered qualitative as well as quantitative. In most of the experiments the current to the electrode E was about twice that to the electrode H; the velocity of the current of air along the cylinder was thus about 0.8 cm. a second.

Comparison of Emanating Power.—The experiments in all cases on the amount of emanation from different substances are comparative. The standard of comparison was usually a sample of 10 grams of thoria as obtained from the maker, which gave out a conveniently measurable quantity of emanation. Preliminary experiments were made to find the connexion between the weight of thoria and the amount of emanation as tested in the cylinder. The following numbers show that the amount of emanation is within the limits of accuracy desired directly proportional to the weight of substance:—

Weight of thoria.	Divisions of scale per second.
2 grams.	1.41
4 ”	2.43
10 ”	6.33
20 ”	13.2

Correction for Natural Leakage.—Even with no emanating material in C the electrometer generally indicates a slight movement on separating the quadrants. This is caused by a small current, chiefly made up of leakage due to conduction over the ebonite, as well as the current produced by the excited radioactivity which has collected on the negative electrode during the course of the day's experiments. It varies from day to day, and is as a rule negligible; but in case of bodies possessing very low emanating power it is necessary to correct for it. The number of divisions of the scale per second indicated by the electrometer-needle when no emanating material is present, is subtracted from the number obtained with the specimen being tested. The corrected number indicates the current due to the emanation alone.

Alternative Method of Comparing Emanating Power.—The apparatus (fig. 1) described in the first paper (Phil. Mag. Sept. 1902) for the comparison of radiations, can also be quite well employed for a comparison of emanating power. In this case, a thick layer of thoria (several grams) is spread over the plate and covered with two thicknesses of ordinary paper.

This has been found almost completely to stop the straight line radiation, whilst allowing the emanation to pass through unimpeded. The current is now measured when a steady state has been reached, due to the accumulation of the emanation. This takes some time, and draughts of air must be guarded against. For this reason, it is less convenient than that first described, but the results obtained by the two methods are almost exactly the same. Thus a sample of "de-emanated" thoria, which gave 12 per cent. of the emanating power of the comparison sample by the first method, gave 13 per cent. by the second method, whilst a sample of oxide prepared from thorium oxalate gave 37 per cent. and 39 per cent. by the two methods respectively. This close agreement in the values by methods so completely different in character is a proof that the indications of the methods are worthy of a great degree of confidence.

III. *The De-emanation of Thoria and the Regeneration of the Emanating Power.*

The emanating power of thoria, as has been stated, is destroyed to a large extent by intense ignition. A closer study of this is the first step in the investigation of the phenomenon. Previous experiments had not succeeded in completely de-emanating thoria, although a reduction to about 15 per cent. of its original value had been accomplished. A sample of this preparation which had been kept for two years had not altered from this value. An experiment was performed in which thoria was heated for one hour by means of a powerful gasoline furnace to the highest temperature which could be safely employed with platinum vessels. The temperature was such that the fireclay walls fused, and the pipeclay of a triangle showed signs of having been softened. It was found that the sample retained about 8 per cent. of its original emanating power. In another experiment, a small platinum crucible filled with thoria was heated for half-an-hour in a small furnace by a large blowpipe and powerful pair of bellows. Some asbestos-wool had completely fused on the outside of the crucible, and the temperature was probably but little lower than in the previous experiment. This sample also retained about 8 per cent. of its emanating power. No further attempt has yet been made to completely destroy the emanating power.

A small quantity of thoria heated in a platinum crucible in the open over an ordinary-sized blowpipe and bellows for five minutes retained about 45 per cent. of its emanating power. The effect of time as well as of temperature was

studied by heating about equal quantities in a platinum crucible over an ordinary Bunsen burner for different periods.

Heated 10 minutes.	Emanating power=61 per cent.
" 1 hour.	" " =59 "
" 24 hours.	" " =42 "

It thus appears that there is a large and practically sudden decrease of emanating power for each temperature above a red heat, followed by a very gradual decrease with time when the temperature is maintained; thus, five minutes on the blowpipe, whilst much more effective than the same time at the temperature of the Bunsen burner, produced rather less effect than 24 hours at the latter temperature.

Effect of Moisture.—The next point to be examined was whether the loss of emanating power could be attributed to the loss of water and desiccation of the thoria by ignition. A sample of de-emanated thoria (retaining about 14 per cent.) was placed in the middle of a Jena glass tube, one end of which was closed and contained water, the other end being drawn out to a jet. This was supported in a powerful tube-furnace in a sloping position, and the part containing the thoria heated to the highest possible temperature, while a slow current of steam from the water at the end was passed over it, escaping by the jet. When all the water was evaporated, the jet was drawn off and the tube allowed to cool in an atmosphere of steam free from air. The thoria, on testing, was found to have been lowered in emanating power to about 7 per cent. The further heating had thus reduced the emanating power without the steam having at all regenerated it.

In the next experiment, the reverse was tried. Two exactly parallel processes were carried out for ordinary thoria possessing the normal amount of emanating power. In the first, it was heated in a porcelain tube in the tube-furnace for three hours, while about 500 c.c. of water were distilled over it from a retort. In the second, another quantity of thoria was heated in exactly the same way for the same time, only a current of well-dried air was substituted for the steam. The result was conclusive: each sample had had its emanating power reduced to exactly the same amount, that is, about 50 per cent. of the original.

These experiments prove that water-vapour exerts no influence either in de-emanating thoria or in effecting a recovery of its lost emanating power.

The Regeneration of the Emanating Power by Chemical Processes.—The task of subjecting de-emanated thoria to a

series of chemical changes to see if it would recover its lost emanating power was then undertaken.

It may first be mentioned that thoria which has been subjected to ignition has changed very materially in chemical and physical properties. The pure white colour changes at temperatures corresponding to the first stages of de-emanation to a light brown, and after subjection to the very highest temperature to a pure pink. At the same time the solubility of the substance in sulphuric acid is greatly diminished. A part always obstinately refuses to dissolve, even after long and repeated boiling with the concentrated acid, although this part is diminished on each successive treatment, and appears to be in no way different from the rest of the substance. No difference, however, occurs in the readiness with which chlorine attacks it when intimately mixed with carbon. The formation of the chloride by this method is the easiest way of dissolving ignited thoria.

Preliminary experiments went to show that emanating power is a quantity which varies, not only with the nature of the chemical compound but also for the same compound very materially with its previous history. Thus the oxide from the oxalate does not possess as a rule so great an emanating power as that used for comparison. The following two exactly parallel experiments were therefore made, the one with the ordinary, and the other with de-emanated thoria possessing 9 to 10 per cent. of the emanating power of the first. Each was converted to chloride in the ordinary way, by mixing with sugar solution, carbonising, and igniting the mixture of oxide and carbon so obtained in a current of dry chlorine. Each sample was then treated with water, the thorium precipitated as hydroxide with ammonia, and the hydroxides washed and dried at 110° . The result was conclusive, for each sample showed the same emanating power. For the first few days after preparation this value increased rapidly, but after having been kept a fortnight both specimens showed about 260 per cent. of the emanating power of the thoria used as a comparison sample.

Thus the process of de-emanating thoria by ignition does not irretrievably destroy the emanating power, for after solution and reprecipitation no difference whatever exists in the emanating power between ordinary and de-emanated thoria. A fair conclusion from these experiments is that the cause of the emanating power is not removed by ignition, but only rendered for the time being inoperative.

IV. *Effect of Conditions upon Emanating Power.*

The experiments just described brought out two new points. Thorium hydroxide possessed an emanating power which increased with time since preparation, and when it attained its maximum it was much greater than that of the oxide. Before any further work was undertaken, it was necessary to make a close study of the influence of conditions upon the emanating power of thorium compounds.

Effect of Temperature.—The effect of increase of temperature on the emanating power of thoria has already been fully investigated by one of us (*Phys. Zeit.* ii. p. 429, 1901). The results, stated briefly, show that an increase in temperature up to a certain limit, in the neighbourhood of a red heat, correspondingly increases the emanating power. At the maximum this is between three and four times that at the ordinary temperature, and is maintained at this increased value for several hours without any sign of diminution with time. When the thoria is allowed to cool, the emanating power then returns to the neighbourhood of the normal value. If, however, the limit of temperature given is exceeded, de-emanation sets in, and even while the high temperature is maintained, the emanating power falls rapidly to a fraction of its former value. On cooling, the substance is found to be more or less de-emanated. It is of interest that no increase of emanating power is observed when de-emanation commences.

These experiments were extended to include the effects of cooling. The platinum tube which contained the thoria was surrounded with a felt jacket containing a mixture of solid carbon dioxide and ether. The emanating power immediately fell to 10 per cent. of its former value. On removing the cooling agent it again rose quickly to nearly the normal.

In another experiment some thoria was surrounded in a platinum crucible with a mixture of solid carbon dioxide and ether, and kept in a vacuum for several hours. On removing it, and allowing its temperature to rise, it possessed much the same value as an ordinary sample, and after standing some time in the air it was again tested, and no difference could be detected between the two.

Thus changes in temperature produce very marked simultaneous changes in emanating power, but between the limits of -110° and an incipient red heat no permanent alteration in the value occurs.

Effect of Moisture.—Dorn (*loc. cit.*) had noticed that moisture produced a moderate increase in the power of thoria

of giving an emanation, and of exciting radioactivity on surrounding surfaces. We have confirmed and extended his results by the following experiments.

Two similar samples of thoria left sealed up for a week, the one in a desiccated atmosphere, the other in air saturated with water-vapour, showed an increase and decrease in emanating power respectively. The moist sample possessed nearly twice as much emanating power as the dry. More complete desiccation, by sealing-up the specimens *in vacuo* with phosphorus pentoxide for a month, did not further reduce the emanating power. Some thoria mixed with concentrated sulphuric acid gave about one half of the usual amount of emanation when vigorously shaken. These experiments show that the presence of water, although producing a marked increase, is not essential for the production of the phenomenon.

Other experiments were made on the effect of light and air on emanating power. The most useful result obtained is that thoria does not change in emanating power when kept in closed vessels under different conditions, but when exposed to the air the emanating power varies within comparatively narrow limits.

Thorium Hydroxide.—This compound, like the oxide, has its emanating power increased by water-vapour. A similar experiment to that described for the oxide gave as the result an emanating power of 400 per cent. of that of thoria for the moist sample and 300 per cent. for the dry. Exposure to the air for a short time again equalized the two values. Carbon dioxide, which thorium hydroxide absorbs from the air to the extent of 2 per cent. of its weight, is without influence on the emanating power.

Effects of Molecular Condition and State of Aggregation of Thorium on the Emanating Power.—Unlike the radioactivity, the emanating power of thorium compounds is by no means mainly controlled by the proportion of thorium present. The effect of temperature in de-emanating thoria and the high value of the emanating power of thorium hydroxide illustrate this. Thorium sulphate, oxalate, and nitrate possess but low emanating power, while thorium carbonate has been obtained with a value five times as great as that of thoria. In general a dense crystalline compound in not very fine powder possesses a much higher emanating power than a light flourey compound in a much finer state of division.

Solution, however, has been found generally to greatly increase the emanating power of soluble thorium salts. In a

careful determination, using 20 grams of finely-powdered thorium nitrate, this worked out to be only 1.8 per cent. of the emanating power of thoria. Dissolved in water, however, and tested for emanation by bubbling a current of air through the solution, it gives about three times as much emanation as thorium oxide. That is, solution in water increases the emanating power of thorium nitrate nearly 200 times. The emanating power, as in the case of solids, is proportional to the weight of substance present, and within the limits tried is not much affected by dilution, for a solution of 10 grams made up to 25 c.c. in volume possessed a similar value when diluted four times.

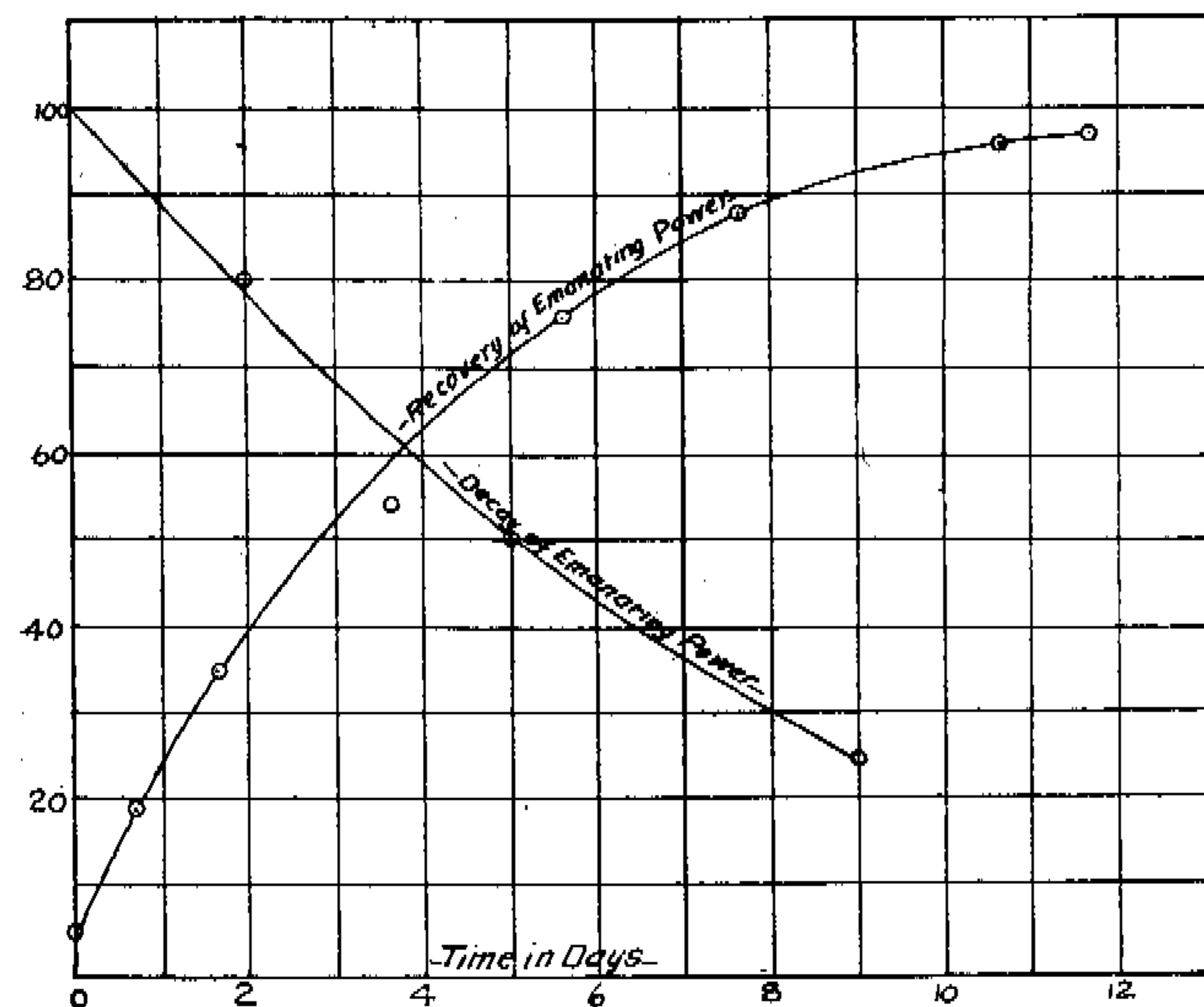
V. The Cause of the Emanating Power of Thorium.

The separation from thorium of ThX, detailed in the first part of this communication, showed that not only the radioactivity but also the emanating power of thorium is connected with the presence of a non-thorium type of matter, ThX. The solutions from which thorium hydroxide had been precipitated by ammonia possessed, when concentrated, about as much emanating power as the solutions from which they were prepared, while the precipitated hydroxide was more or less completely de-emanated. On allowing these preparations to stand, the emanating power of the filtrates gradually disappeared, while that of the hydroxide in most cases rose steadily with time, till at the end of a fortnight they had attained a maximum between three and four times that of ordinary thoria. This recovery of the emanating power in the case of the hydroxide was noticed long before the similar change of its radioactivity was observed, but the two phenomena admit of a similar explanation. If, in the precipitation by ammonia, care is taken to remove the ThX completely, the thorium hydroxide is at first almost devoid of emanating power. The small fraction that remains—only a few per cent. of the maximum—can be accounted for by the reproduction of ThX during the time taken to dry the precipitate.

The Rate of Recovery and Decay of Emanating Power.—The rate of decay of the emanating power of ThX, and the recovery of this property by the thorium from which it had been separated, were then investigated in parallel with the similar experiments on radioactivity already described. One quarter of the concentrated filtrate used for the latter purpose was taken, and the decrease of its emanating power with time measured. The increase of emanating power of the thorium hydroxide from which it had been prepared was also

measured. Fig. 2 expresses the results. The decay-curve is merely approximate, for it is not easy to accurately take the emanating power of a liquid without special arrangements to assure the constancy of the air-current and the shaking of the solution.

Fig. 2.



The experiments, although only of a preliminary character, bear out the conclusion that emanating power decays and recovers according to the same law and at the same rate as the radioactivity of ThX, and that it is therefore one of the properties of the latter and not of thorium. The decay-curve given, so far as it can be relied upon, shows that the emanating power of ThX at any instant is proportional to its radioactivity.

VI. The Chemical Nature of the Emanation.

The following work has reference to the emanation itself, and not to the material producing it, and was designed to see whether the emanation possesses chemical properties which would identify it with any known kind of matter. It had been noticed at the time of its discovery that it passed unchanged through concentrated sulphuric acid. The same holds true of every reagent that has been investigated.

The effect of temperature was first tried. The air containing the emanation, obtained in the usual way by passage over thoria, was led through the platinum tube heated

electrically to the highest attainable temperature, and also through the tube cooled by solid carbon dioxide and ether. The tube was then filled with platinum-black, and the emanation passed through it in the cold, and with gradually increasing temperatures, until the limit was reached. The effect of the intense heat was to convert the platinum-black completely into platinum-sponge. In another experiment the emanation was passed through a layer of red hot lead-chromate in a glass tube. The current of air was replaced by a current of hydrogen, and the emanation sent through red hot magnesium-powder and red hot palladium-black, and, by using a current of carbon dioxide, through red hot zinc-dust. In every case the emanation passed without sensible change in the amount. If anything, a slight increase occurred, owing to the time taken for the gas-current to pass through the tubes when hot being slightly less than when cold, the decay *en route* being consequently less. It will be noticed that the only known gases capable of passing in unchanged amount through all the reagents employed are the recently-discovered members of the argon family.

But another interpretation may be put upon the results. If the emanation were the manifestation of excited radioactivity on the surrounding atmosphere, then since from the nature of the experiments it was necessary to employ in each case, as the atmosphere, a gas not acted on by the reagent employed, the result obtained might be explained. Red hot magnesium would not retain an emanation consisting of radioactive hydrogen, or red hot zinc-dust an emanation consisting of radioactive carbon dioxide. The correctness of this explanation was tested in the following way. Carbon dioxide was passed over thoria, then through a T-tube, where a current of air met and mixed with it, both passing on to the testing-cylinder. But between this and the T-tube a large soda-lime tube was introduced, and the current of gas thus freed from its admixed carbon dioxide before being tested in the cylinder for emanation. The amount of emanation found was quite unchanged, whether carbon dioxide was sent over thoria in the manner described, or whether an equally rapid current of air was substituted for it, keeping the other arrangements as before. The theory that the emanation is an effect of the excited activity on the surrounding medium is thus excluded. It is *a priori* improbable on account of the very different rates of decay of the activity in the two cases. The interpretation of the above experiments must therefore be that the emanation is a chemically inert gas analogous in nature to the members of the argon family.

In light of these results, and the view that has already been put forward of the nature of radioactivity, the speculation naturally arises whether the presence of helium in minerals and its invariable association with uranium and thorium may not be connected with their radioactivity.

VII. *The Nature of Emanating Power.*

The foregoing results therefore find their simplest expression on the view that, just as a chemical change is proceeding in thorium whereby a non-thorium material is produced, so the latter undergoes a further reaction, giving rise to a gaseous product which in the radioactive state constitutes the emanation.

It will be seen at once that this secondary change is of a different kind from the primary, for it is affected apparently by the conditions in a very marked manner. It was shown that moisture, the state of aggregation, and temperature influenced the value of the emanating power. From -80° to a red heat the latter regularly increases in the ratio of 1:40 in the case of thorium oxide, while the ratios between the values for thorium nitrate in the solid state and in solution is as 1:200. The secondary reaction appears therefore at first sight much more nearly allied to ordinary chemical reaction than the primary. It must not be forgotten, however, that the laws controlling the manifestation of the two phenomena—radioactivity and emanating power—are of necessity very different. In the former we deal with the intensity of radiations emitted by a solid, in the latter with the rate of escape of a gas into the surrounding air from either a solid or a liquid. Since this gas is detected by its radioactivity, and this decays extremely rapidly with time, a very slight delay in the rate of its escape will enormously affect the experimental value obtained for emanating power.

On the other hand, it is now well established by experiment that sometimes thorium compounds de-emanated chemically by removal of ThX do not recover their normal emanating power with time, but remain constant at a lower value. On one occasion a carbonate was prepared which possessed hardly any emanating power until it was again dissolved and precipitated. In another experiment two samples of hydroxide prepared from different nitrates were tested together for rise of emanating power. That of the one rose normally to its maximum (as in fig. 2), which was twenty times the minimum. The other started from the same minimum, but rose to a maximum only one-fourth as great. When the experiment

was repeated under the same conditions, using the same sample of nitrate, the compound behaved normally. It thus appears that the emanation can be almost entirely prevented from escaping in the radioactive state in some cases, and partially prevented in others, where no visible peculiarity of physical condition exists, and where other preparations similarly prepared behave normally.

These are outstanding points in the theory which remain to be explained. It is not possible at present to decide whether these variations of emanating power are caused by an alteration in the velocity of the reaction which produces the emanation, or by an alteration in the time taken for the latter to escape. The experiments detailed in the first paper on the augmentation of the proportion of excited activity in compounds de-emanated by ignition appear to favour the view that the change still proceeds, but the emanation does not succeed in escaping. The experiment on the regular variation of emanating power with temperature might be explained quite well by either hypothesis.

VIII. *The Excited Radioactivity from Thorium.*

Since the emanation gives rise to the phenomenon of excited radioactivity, and the latter appears to be caused by an intensely active invisible deposit of matter, it must be supposed that a tertiary change is taking place. The emanation, a gaseous product of the secondary reaction, is again changing and giving rise to a third reaction-product causing the excited activity. The fact that it is manifested entirely on the negative electrode in an electric field, points to the positive ion being the means by which it is transported. Without, in the present paper, going further into the consideration of excited radioactivity, it may be mentioned that the successive changes occurring in the thorium atom are not yet ended at this stage. The fact that the excited radiation consists in part of cathode-rays may be recalled here. Further, the intensity of excited activity at first *increases* from the time of its formation, exactly as in the case of ThX newly separated from thorium, the increase reversing the effect of the normal decay. The radium excited activity behaves in a somewhat analogous manner. The matter in this case causing excited activity does not appear to be homogeneous, but behaves in its action towards acids, &c. as if consisting of two different kinds (compare Rutherford, *Phys. Zeit.* p. 254, 1902).

IX. *Further Theoretical Considerations.*

Enough has been brought forward to make it clear that in the radioactivity of thorium, and, by analogy, of radium, we are witnessing the effect of a most complex series of changes, each of which is accompanied by the continuous production of a special kind of active matter. The complexity of the phenomenon gives rise to an important question concerning the fundamental relation between the changes which occur and radioactivity. So far it has been assumed, as the simplest explanation, that the radioactivity is *preceded* by chemical change, the products of the latter possessing a certain amount of available energy dissipated in the course of time. A slightly different view is at least open to consideration, and is in some ways preferable. Radioactivity may be an *accompaniment* of the change, the amount of the former at any instant being proportional to the amount of the latter. On this view the non-separable radioactivities of thorium and uranium would be caused by the primary change in which ThX and UrX are produced. The activity of ThX would be caused by the secondary change producing the emanation, the activity of the emanation by a tertiary change in which the matter causing the excited activity is produced, the activity of the latter being derived from still further changes. The law of the decay of the activity with time (equation 1 first part) in all cases but the primary then appears as the expression of the simple law of chemical change, in which one substance only alters at a rate proportional to the amount remaining. In the primary change the amount remaining is infinitely great compared with the amount that alters in short time, and therefore the velocity of reaction is constant. This view certainly affords an explanation of why the emanating power of ThX is proportional to the radioactivity. So long as the latter is considered a consequence of what has occurred there is no reason why this should be so. But if it is considered the accompaniment of the change in which the emanation is formed the result follows naturally. Further and more exact determinations of the rate of rise and decay of emanating power are therefore called for.

In the case of uranium the changes so far as they can be followed by the radioactivity appear to be at an end with that which causes the activity of UrX. It is of interest that this substance gives only cathode-rays, and that it continues to do so for many weeks after its separation from uranium. This gives rise to the question whether any connexion can be established between the nature of the radiation and the kind of change producing it.

The only consideration which is opposed to this view is the existence of polonium. The radiations of this body resemble closely the non-separable radioactivity of uranium, both in penetrating power and the absence of deviable rays. But all attempts (Soddy, *loc. cit.*) have so far failed to separate polonium from uranium, and until this is done its existence does not of itself affect the present question.

It seems as if a more satisfactory explanation of the residual activities common to both uranium and thorium, and of the connexion between the emanating power and radioactivity of ThX, is obtained on the modified view. But further work, both on this latter point and on the nature of polonium, must be awaited before the connexion between radioactivity and chemical change can be considered exactly determined.

Macdonald Physics Building,
Macdonald Chemistry and Mining Building,
McGill University, Montreal.

LXV. *On the Conditions necessary for Equipartition of Energy.*
By J. H. JEANS, B.A., Isaac Newton Student and Fellow
of Trinity College, Cambridge*.

Introduction.

§ 1. **T**HE object of the present paper is, firstly, to give a proof of Boltzmann's Theorem on the Equipartition of Energy from a somewhat new point of view; and, secondly, to examine what are the precise conditions under which equipartition will take place, and whether these conditions are such as will occur in an actual gas.

At the outset it must be explained that the equations leading to the law of distribution admit of a simple hydrodynamical interpretation in generalized space of n -dimensions. I have made use of the hydrodynamical analogy for two reasons. In the first place it is easier to think in terms of generalized space than in terms of multiple integrals; and in the second place the terminology and results of hydrodynamics being ready to hand, a great deal of obscurity and repetition may be saved by starting at once from the hydrodynamical standpoint. So long as we only use hydrodynamical results and conceptions which have a mathematical (as opposed to a physical) basis, there will be no danger of a faulty "argument by analogy."

* Communicated by the Author.