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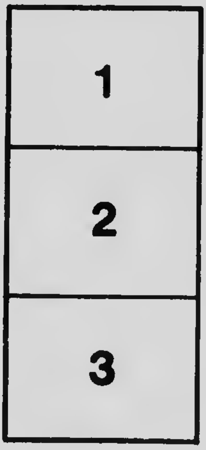


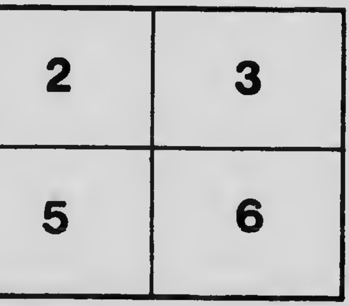

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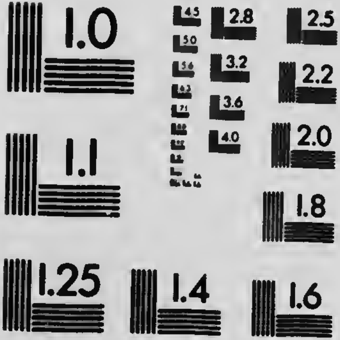

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No. 4. A RADIOACTIVE GAS FROM CRUDE PETROLEUM, BY E. F. BURTON.



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# A RADIOACTIVE GAS FROM CRUDE PETROLEUM

BY

E. F. BURTON, B.A.

## A RADIOACTIVE GAS FROM CRUDE PETROLEUM

In the course of their investigations on the radioactivity of the atmosphere Elster and Geitel[1] have shown that the soil and rock-masses constituting the surface layers of the earth are the source of an emanation, or gas, which gradually escapes into the air, and there exhibits properties analogous to the radioactive emanations from thorium and radium. In a conjoint paper by Professor McLennan and myself[2] on the conductivity of air confined in receivers of different metals some observations are cited which indicate that metals generally are, to a slight degree, the source of a similar emanation. This result has since been confirmed by Strutt,[3] who found that air drawn through a glass tube heated just below redness and containing scrap copper acquired a conductivity three or four times its normal value. Strutt[3] has also shown that a highly radioactive emanation can be obtained by bubbling air through mercury heated to about 300°C. More recently Professor J. J. Thomson[4] established the existence of a radioactive gas in the Cambridge tap-water, as well as in the water from a number of wells in different parts of England. Similar results have been obtained by Himstedt[5] at Freiburg, and by Lord Blythswood and H. S. Allen[6] with the mineral waters of Bath. Later still Adams[7] made a careful study of the radioactive gas in Cambridge tap-water, and his results, as well as those of Strutt on the emanation from mercury, go to show that the activity in all these cases is due to the presence of a substance very similar to, if not identical with, the emanation from radium.

In the following paper an account is given of some experiments with a highly radioactive gas obtained from crude petroleum, which, both in the rate at which its activity decays and

[1] Phys. Zeit., 3 Jahr. 24, p. 574. Denkschr. d. Kommission fur luftelect. Forschungen (München, 1903).

[2] Phil. Mag., 5th series, June, 1903, p. 699.

[3] Phil. Mag., 6th series, July, 1903, p. 113.

[4] Proc. Camb. Phil. Soc. xii, 3, 1903, p. 172.

[5] Berichte der Naturf. Ges. von Freiburg i. B., 1903, xiii, p. 101.

[6] Nature, Jan. 14, 1904, p. 247.

[7] Phil. Mag., 6th series, November, 1903, p. 563.

in the nature of the induced radioactivity it produces, very closely resembles the emanations dealt with by the investigators just mentioned.

*Apparatus.*—The petroleum used in the experiments was obtained from one of the wells belonging to Mr. A. C. Edward, of Petrolia, Ontario, to whom my most sincere thanks are due for many samples of oil supplied during the course of the investigation. The petroleum from this locality is drawn directly from the corniferous limestone which lies at a depth of four hundred and sixty-five feet below the surface, and, while it may possibly originate in these rocks, there are reasons for concluding that the oil has its source in a deeper stratum, very probably in the underlying Trenton formation.

The petroleum to be tested was contained in a large three-litre flask, D (Fig. 1), supported in a water bath. This flask was connected with a wash bottle, E, partly filled with concentrated sulphuric acid, and to a second flask, F, embedded in ice for the purpose of condensing any vapours from the heated oil. The tube, S, was filled with phosphoric pentoxide, and the tube, H, tightly packed with glass wool. The vessel, A, made of thin galvanized iron, 62 cms. long and 25 cms. in diameter, was provided with an exploring electrode, C, which was supported by an ebonite plug carrying a guard tube, B. The rod, C, was connected to one of the pairs of quadrants of a quadrant electrometer of the Dolzaleck type, whose sensitiveness was such that a potential difference of one volt between the quadrants gave a deflection of 1,100 mms. on a scale at a distance of one metre. Throughout the experiments the cylinder, A, was maintained at a potential of 168 volts by a battery of small storage cells, and the conductivity of the gas which it contained was determined by measuring the saturation current to the exploring electrode. This saturation current when the cylinder, A, was filled with ordinary dry air was about 16.5 scale divisions per minute. After heating the water in the bath to the boiling point, air was bubbled for fifteen minutes through the oil and drawn into the cylinder, A, by means of a water pump. The cylinder was then disconnected from the tube, H, and hermetically sealed, after which measurements were made, from time to time, on the conductivity of the gas which it contained. The density of this gas was determined in every case, and found to be about 1.05, air being taken as unity.

*A Radioactive Emanation.*—On first introducing into the cylinder the air which had passed through the oil, it was found to have an initial conductivity very greatly in excess of that of normal air. Its conductivity steadily increased, after the cylinder was closed, for about three hours, when it reached a maximum value, after which it slowly decreased approximately in a geometrical progression with the time. Fresh air passed through different samples of petroleum into the cylinder under exactly similar conditions was found to possess different initial conductivities, but, in every case, the conductivity of the confined air steadily rose in about three hours to a maximum about 40 per cent. in excess of the initial value. It then decayed according to an exponential law, always dropping to one-half value in about 3.125 days. A typical set of observations on the conductivity of air bubbled through one of the samples of oil is given in Table I, the time being reckoned from the moment when the cylinder was closed.

TABLE I.

| Time. Hrs. | Min. | Current: Arbitrary Scale. | Time. Hrs. | Min. | Current: Arbitrary Scale. |
|---|---|---|---|---|---|
| .. | 10 | 92 | 27 | .. | 92 |
| .. | 30 | 95.6 | 41 | 30 | 83.5 |
| 1 | 4 | 103 | 50 | .. | 77.8 |
| 1 | 35 | 111.7 | 67 | .. | 71 |
| 2 | 8 | 116.5 | 73 | 30 | 67.7 |
| 2 | 43 | 119.7 | 95 | .. | 60.3 |
| 9 | 30 | 111.6 | 116 | 30 | 55.5 |
| 20 | .. | 101 | 128 | .. | 50.8 |
| 23 | .. | 95.7 | 138 | 30 | 48.6 |

These results are shown graphically in Fig. II, where the ordinates of the curve represent the conductivity of the gas, and the abscissæ the times in hours.

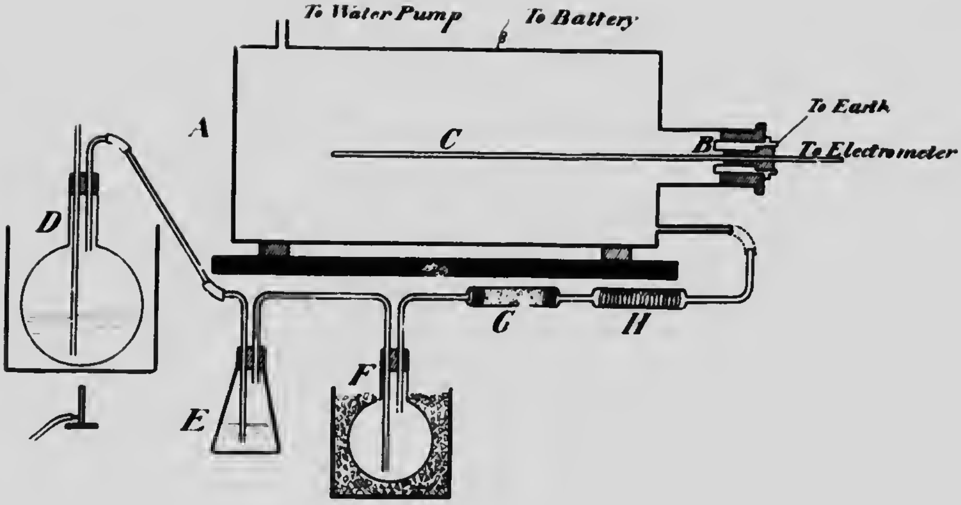
As in the experiments of Professor Thomson with the Cambridge tap-water and those of Strutt with mercury, all of the observed phenomena lead to the conclusion that the air, in passing through the petroleum, becomes mixed with some radioactive gas or emanation. The initial portion of the curve leading up to the maximum corresponds exactly to that of the curve given by Rutherford[1] for the emanation from radium, and also to that of the curve given by Strutt for the radioactive gas obtained by bubbling air through mercury, and may be explained in the same way. The value of the conductivity immediately after the cylinder has been sealed measures the ionization due to the emanation itself. But, according to the disintegration theory proposed by Rutherford, the emanation is continuously producing by its decay the matter which causes excited radioactivity, and the ionizing power added by this latter material more than neutralizes, for a time, the decrease due to the decay of the emanation. Thus the conductivity of air freshly charged with this emanation gradually increases to a maximum state, which is reached when the loss in the ionizing power due to the decay of the emanation is just equalled by the gain contributed by the excited radioactivity produced in this process of decay.

From this time the rate of change indicated gives the rate of decay of the emanation. The law which the rate of decay of the emanation from radium follows may be expressed by the equation:

$$I_t = I_0 \, e^{-\lambda t}$$

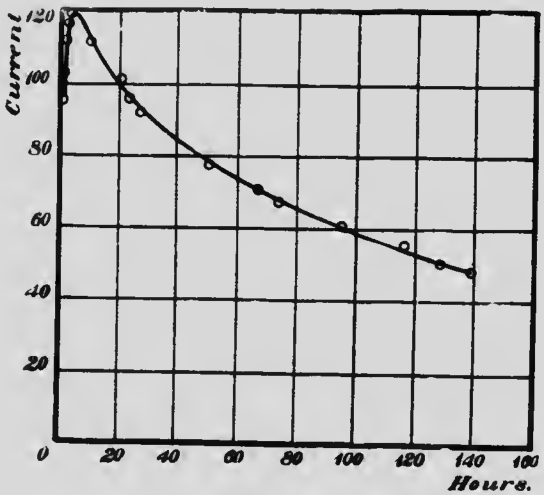
where $I_0$ is the value of the conductivity at any given time, $I_t$ the value after an interval of t seconds, e the base of natural logarithms and $\lambda$ a constant. By using this equation the values of $^1/\lambda$ have been determined for a number of pairs of the readings given above and the results are tabulated in Column I of Table II. These values of $^1/\lambda$, which give a mean of 557,000, show a marked increase with the time, and consequently indicate that the rate of decay is slower than that required by the law given above. This departure from the law of decay is probably due to a slight trace of a more persistent radioactive substance in the gas than the emanation and will be referred to later.

[1] Phil. Mag., 5th series, April, 1903, p. 445.



To Water Pump
To Battery
To Earth
To Electrometer
A
B
C
D
E
F
G
H


Fig. 1.



Current
120
100
80
60
40
20
0
20
40
60
80
100
120
140
160
Hours.


Fig 2.

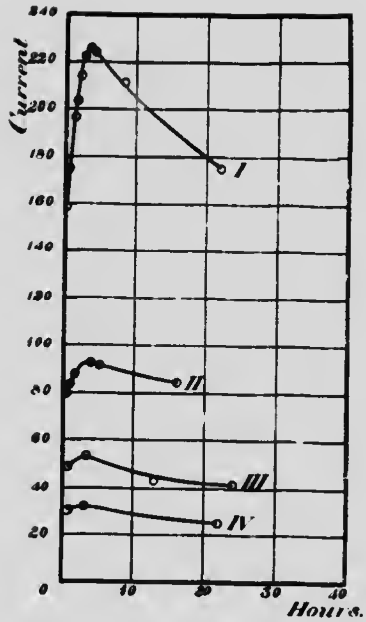

Current
240
220
200
180
160
140
120
100
80
60
40
20
0
10
20
30
40
Hours.
I
II
III
IV

**Fig. 3.**

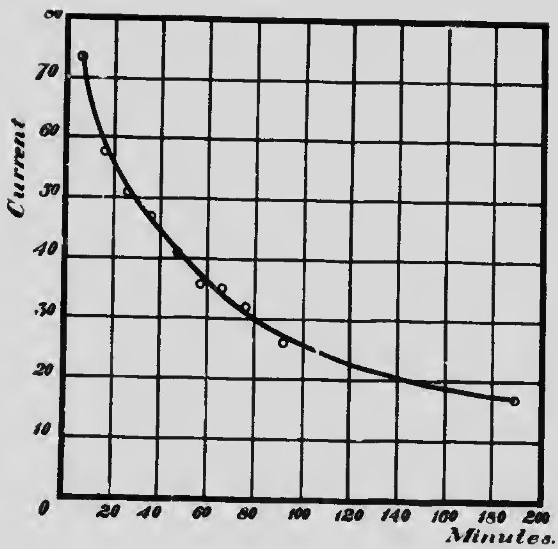

Current
80
70
60
50
40
30
20
10
0
20
40
60
80
100
120
140
160
180
200
Minutes.

**Fig. 4.**

TABLE II.

| Column I. Burton. | | | Column II. Strutt. | | | Column III. Adams. | | |
|---|---|---|---|---|---|---|---|---|
| Time in Hrs. | Current: Arb. Scale. | $\frac{1}{\lambda}$ | Time in Hrs. | Current: Arb. Scale. | $\frac{1}{\lambda}$ | Time in Hrs. | Current: Arb. Scale. | $\frac{1}{\lambda}$ |
| 0 | 119.7 | 360,000 | 0 | 140 | 379,000 | 0 | 188 | 366,000 |
| 17 | 101 | 414,000 | 18 | 118 | 389,000 | 16.7 | 160 | 401,000 |
| 47 | 77.8 | 669,000 | 42 | 94.5 | 472,000 | 40.4 | 120 | 494,000 |
| 64 | 71 | 617,000 | 66 | 78.7 | 504,000 | 64.8 | 108 | 381,000 |
| 92 | 60.3 | 726,000 | 90 | 66.3 | 371,000 | 88.9 | 86 | 372,000 |
| 135.6 | 48.6 | | 140.5 | 40.6 | | 139.6 | 53 | 573,000 |
| | | | | | | 160.8 | 46 | |
| $\frac{1}{\lambda}$ = 557,000 | | | $\frac{1}{\lambda}$ = 423,000 | | | $\frac{1}{\lambda}$ = 425,000 | | |
| Half value in 3.125 days. | | | Half value in 3.18 days. | | | Half value in 3 days. | | |

In Column II of Table II is given a set of Strutt's readings for the ionization due to the radioactive gas in mercury, and in Column III the values obtained by Adams with the active emanation in Cambridge tap-water. The calculated values of $^1/\lambda$ are inserted in both cases, but do not show the increase exhibited by the numbers in Column I. The averages of the three series of values of $^1/\lambda$ given in Table II, together with the mean values of the same constant obtained by Mme. Curie[1] and by Rutherford[2] for the decay of the emanation from radium, as well as the mean value calculated from Himstedt's results for the radioactive gas in water are collected in Table III. The values-show a very close agreement, and lead to the conclusion that the active gases from petroleum, spring water, and mercury are very probably identical with the emanation from radium.

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1. Thèses prés. à la Faculté des Sci. de Paris, 1903.
2. Phil. Mag., 5th series, April, 1903, p. 445.

TABLE III.

| Experimenter. | Source of Emanation. | Value of $\frac{1}{\lambda}$ |
|---|---|---|
| Mme. Curie | Radium | 497,000 |
| Rutherford | Radium | 463,000 |
| Strutt | Mercury | 423,000 |
| Adams | Tap-water | 425,000 |
| Himstedt | Water | 491,000 |
| Burton | Petroleum | 557,000 |

In his experiments with the water from the Cambridge mains Professor J. J. Thomson found that when the water had once been well boiled the gas expelled on any subsequent re-boiling was not appreciably radioactive. In the present investigation air was drawn through a selected sample of oil into the cylinder on three consecutive days and again on the sixth day, the first measurement being made about 24 hours after the petroleum had been pumped from the well. Each time the oil was used the bath was brought up to the boiling point and the air bubbled through it for 15 minutes, when observations on the conductivity of the air in the cylinder were commenced and continued at intervals over a period of about 20 hours.

TABLE IV.

| Curve I. | | | Curve II. | | | Curve III. | | | Curve IV. | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Time. H. | Time. M. | Current: Arb. Sc. | Time. H. | Time. M. | Current: Arb. Sc. | Time. H. | Time. M. | Current: Arb. Sc. | Time. H. | Time. M. | Current: Arb. Sc. |
| | 10 | 158.7 | | 30 | 80 | | 40 | 49 | | 35 | 29.8 |
| | 30 | 174.2 | 1 | | 83.4 | 3 | 10 | 53 | | 55 | 30.3 |
| 1 | 5 | 196.7 | 1 | 30 | 87.2 | 13 | | 43 | 3 | | 31.6 |
| 1 | 30 | 203.7 | 3 | 40 | 92.6 | 24 | | 41.5 | 22 | | 25.2 |
| 1 | 50 | 214.2 | 5 | | 92 | | | | | | |
| 2 | 30 | 222.5 | 16 | | 84.3 | | | | | | |
| 3 | 30 | 226 | | | | | | | | | |
| 4 | | 224.2 | | | | | | | | | |
| 8 | 15 | 211.1 | | | | | | | | | |
| 22 | | 176 | | | | | | | | | |

The results, which are embodied in Table IV and illustrated by the curves in Fig. III, show that the activity acquired by fresh air when drawn through the oil gradually decreased from day to day. The curves corresponding to the different tests exhibit the same characteristics as that in Fig. II. In each case the conductivity rose to a maximum in about three hours and then gradually decreased. The maximum currents in the four trials were respectively 13.9, 5.6, 3.2, and 1.9 times the conductivity of the ordinary air, thus showing that the oil at the end of a week still possessed in a marked degree the power to impart radioactivity to air drawn through it. Experiments made with a sample of oil which had been used in some preliminary tests and had been placed aside in a tightly corked glass vessel for over a month gave values almost identical with those represented by Curve IV, Fig. III, the maximum conductivity impressed in this case being 1.6 times that of the normal air. From these results it would appear that there is present in crude petroleum an active substance more persistent than the emanation from radium, perhaps a minute quantity of radium itself. If this be so, the air drawn through the oil might possibly carry with it into the cylinder a slight trace of this substance. Such a condition would explain the departure from the law of decay $I_t = I_o e^{-\lambda t}$ exhibited by the increasing values of $1/\lambda$ in Column 1 of Table II.

*Induced Radioactivity.*—Each time the gas containing the emanation was blown from the cylinder the conductivity of the ordinary air admitted from the room was found to be still very high. Repeated tests showed that the initial conductivity of this fresh air was about 35 per cent. of that of the displaced gas, but in every case it quickly fell, until after about two hours the conductivity reached the normal value of 16.5. In expelling the emanation a blast of air was sent through the cylinder continuously for five minutes by means of a small foot-pump, after which the receiver was again sealed.

Measurements were then made on the conductivity at short intervals, and in Table V are given the results of one of these tests, the time being taken from the closing of the cylinder. In this particular case, the cylinder while filled with the air con-

TABLE V.

| Time in Minutes. | Current: Arbitrary Scale. |
|---|---|
| 5 | 73.8 |
| 15 | 58.2 |
| 25 | 50.6 |
| 35 | 47.2 |
| 46 | 41 |
| 56 | 35.6 |
| 65 | 35.4 |
| 75 | 32 |
| 91 | 26 |
| 200 | 16.7 |

taining the emanation was maintained at a negative potential of 168 volts for 22 hours and during this time the conductivity rose from its initial value of 158.7 to a maximum of 226 and then fell to 176.3 before the expulsion took place.

The curve given in Fig. IV, in which the ordinates represent currents and the abscissae times, illustrates the results in this table. From this curve it is seen that the conductivity decreases in a geometrical progression with the time, falling to one-half value in about 35 minutes. This phenomenon is exactly analogous to that which other investigators have found in working with the radioactive emanations from thorium and radium and which has been explained on the assumption that these emanations have but a transitory existence and are gradually transmuted to a new substance which has a definite rate of of decay and which is the cause of the so-called induced or excited radioactivity. On this view it is clear that, from the observations above, the active emanation from petroleum also produces the substance which is responsible for induced radioactivity, and that the presence of this substance in the cylinder is the cause of the high conductivity of the fresh air which replaced that blown out.

An experiment giving similar results was conducted under the same conditions as that just described, except that the cylinder was maintained for 22 hours before the emanation was expelled at a positive potential of 168 volts. This would show that the substance responsible for excited radioactivity was left in the cylinder in both cases when the air was blown out and, as it is known that negatively charged conductors in the presence of radioactive emanations become more active than those positively electrified, it is very probable that in the first experiment the excited radioactivity was deposited on the walls of the receiver, while in the second case it was concentrated upon the electrode, C.

A confirmation of this conclusion was obtained by exposing a conductor under negative electrification, and then under positive, to the petroleum emanation. The exploring electrode, C, was taken from the cylinder, A, and suspended in a large glass tube, through which air containing the radioactive emanation was drawn. It was connected for half an hour with the negative terminal of an electrical machine giving a potential of about 10,000 volts, and on being replaced in the receiver it increased the conductivity of the air to about three times its normal value. The conductivity in this case fell to a half value in the same time as before. When the exploring electrode was suspended under a positive electrification of 10,000 volts, for the same time, in the current of air containing the emanation, it did not acquire any appreciable activity.

It has been shown by Mme. Curie, Rutherford and others that the induced radioactivity from the radium emanation decays to one-half value in about thirty minutes, and Adams has found that the induced radioactivity from the gas in Cambridge tap-water falls to half value in about thirty-five minutes. These values are practically the same as that determined in the present investigation, and confirm the conclusion already arrived at that the active gas from crude petroleum is very probably identical with the emanation from radium.

*Conclusions.*—Summarizing the results given in the foregoing paper we have the following:

1. Fresh crude petroleum has been found to contain a strongly radioactive gas which is similar in its rate of decay, and also in the rate of decay of the induced radioactivity which it produces, to the emanation from radium and to the emanations obtained by a number of experimenters from mercury and from certain waters fresh from the earth.

2. This radioactive gas decays approximately according to an exponential law, falling to a half value in 3.125 days.

3. It produces an induced radioactivity whose rate of decay is such that it falls to a half value in about 35 minutes.

4. There are indications of the existence in crude petroleum of slight traces of a radio-active substance more persistent than the radium emanation.

In a paper published during the progress of the experiment by Elster and Geitel[1] reference is made to a recent investigation by Himstedt on the radioactivity of petroleum, but up to the present time this communication has not been received, so that a comparison with his results cannot be made here.

In conclusion, I desire to express my thanks to Professor J. C. McLennan for suggesting the research and for his invaluable aid and advice at all times. I also wish to acknowledge my indebtedness to Mr. L. Gilchrist and to Mr. S. Dushman for kindly aiding me in making some of the observations.

[1] Archives des Sci. Phys. et Nat., 4. t. XVII, Jan. 1904, pp. 5-22.

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