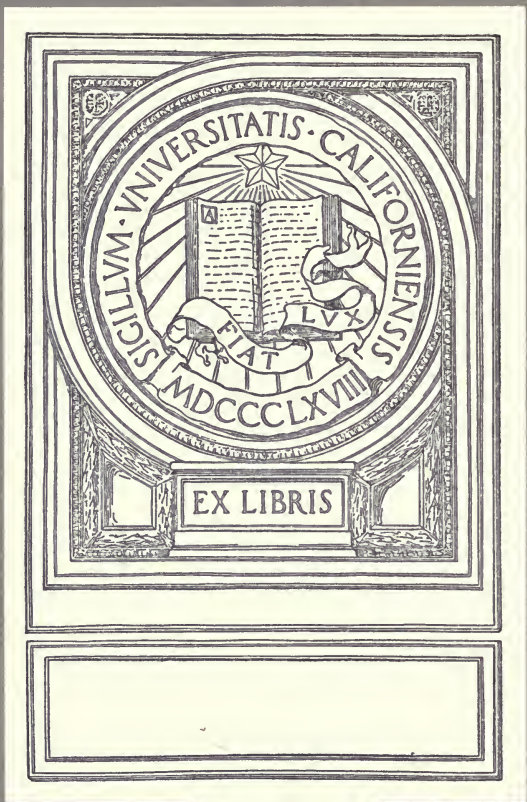


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# RADIUM AND RADIO-ACTIVE SUBSTANCES

Their Application Especially to Medicine .

BY

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TO

ERNEST RUTHERFORD

WHOSE INVESTIGATIONS ON RADIO-ACTIVITY  
ARE WORTHIER OF A HIGHER TRIBUTE



## PREFACE.

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To fill a demand for an inexpensive non-mathematical work on the subject of radium and its application in medicine, arrangements were made for the publication of this book. At that time the excellent treatises of Rutherford and Soddy had not appeared. To anyone purposing the prosecution of investigations, these works are indispensable. One treats the phenomena of radio-activity from the point of view of a physicist, while the other looks at them more with the eyes of a chemist. The appearance of these works has made it necessary to alter this book somewhat. The technical details have been given so admirably by these co-laborers that it has been thought just as well to omit much here. This book emphasizes a phase naturally but hinted at by them, namely, the application of radio-active substances in medicine. The writer is not in a position to harmonize the contradictory evidence given in reputable medical journals as to the therapeutic uses of the salts of radium, consequently the observations have been impartially reported. Physicians of prominence, who have had much to do with the use of this novel substance in their practice, have been good enough to revise the chapter bearing upon that phase of the subject.

It has been deemed advisable for comparison to annex a short chapter on other therapeutic radiations.

As many physicians will have neither the time nor the opportunity to study the larger works, sufficient of the general subject has been presented for a fairly clear conception of our present knowledge of these startling, perhaps revolutionary, phenomena.



Although the work makes no pretense at completeness, all known sources of information have been freely drawn upon. In most cases due credit has been given.

The bibliography, which was prepared, has been omitted. A most complete index to the extensive literature of radium by Dr. George F. Kunz is in press for the United States Geological Survey.

Dr. Fritz Zerban and Mr. Frederick E. Breithut have generously followed the proof-sheets. Mr. N. R. Graham was good enough to prepare the index.

New York, 1905.

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## Radium and Radio-Active Substances.

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### CHAPTER I.

#### THE PHENOMENON OF RADIO-ACTIVITY.

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If there be one thing which may be said to characterize science and its progress, it is evolution, or growth. Practically all the great movements of science and its modern marvels are linked to the past. The phenomenon of radio-activity, which has astonished a civilization accustomed to wonders, is no exception.

Without going too far back, attention may be called to the now well known fact that an electric spark passes through the air in a zigzag line, the length of which varies with the distance between the charged and uncharged bodies. The intensity of the spark varies with the charge, a corresponding difference being observed, as when the hand is passed over a cat's fur or surcharged clouds relieve themselves in a violent flash of lightning. The discharge presents a very different appearance, however, when the air is rarified, as originally investigated by Gassiot.

Geissler, of Bonn, was the first to imprison gases under diminished pressure in tubes provided with electrodes, that is, conducting terminals by which the discharge may be carried in or cut. Geissler tubes are so exhausted that there exists an internal pressure of about one-thousandth of an atmosphere. The discharge, visibly passes (Fig. 1) between the anode (positive) and cathode (negative), the one terminal being of as

much importance apparently as the other. Plücker has shown that the color of the light produced is not dependent upon the substance of the electrodes. It varies with the nature of the gas or vapor, being crimson with hydrogen and purple-red with nitrogen, and so on.

In 1876 Crookes made an elaborate investigation of the phenomena produced by the electric discharge in much higher vacua<sup>1</sup>. The pressure within the Crookes tubes is about one thousandth that of the Geissler tubes, or one-millionth of an atmosphere, and the path of the discharge is no longer visible. The discharge is independent of the anode and appears to proceed from the cathode alone (Fig. 2). Further, the luminous

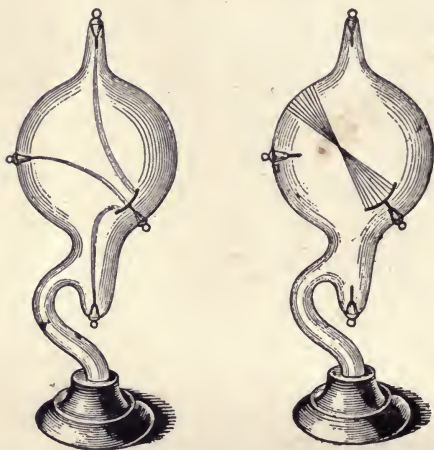


Fig. 1.

Fig. 2.

Fig. 1. In a Geissler tube the discharge visibly passes between anode and cathode, the location of the former being of little importance.

Fig. 2. In a Crookes tube the cathode rays are projected in straight lines from its surface; their presence is noted by the fluorescence of the walls of the tube opposite the negative terminal.

effect upon the glass is directly opposite the cathode, which indicates that these rays move in straight lines and strike upon the glass exactly opposite that electrode. With the Geissler

1. Phil. Trans. CLXX, 135, 641; Nature XX, 419, 436.

tubes, on the other hand, the discharge may be made to follow devious paths, depending upon the shape of the apparatus, thus producing exquisite effects. (Fig. 3).

The production of light without any considerable heat by the action of such rays was designated *luminescence* by E. Wiedemann. The color of the light depends not upon the gaseous contents of the tubes, but the solid material upon which the rays impinge. Some diamonds, for example, glow splendidly white, while coral gives an intensely brilliant reddish or purplish luminescence

Two other interesting properties of the cathode rays must be mentioned. A tube was prepared by Crookes (Fig. 4),

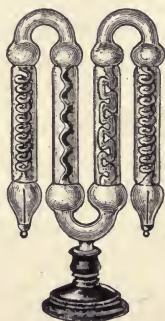


Fig. 3.

Fig. 3. In a Geissler tube the current may pass devious paths.



Fig. 4.

Fig. 4. A Crookes tube provided with a window in an aluminum-shield and fluorescent screen. The cathode rays projected, on the passage of the current, as a beam upon the screen, appear as a ribbon of light indicated by the shading. On bringing the pole of a strong magnet above, the cathode rays are attracted or repelled according to the polarity. The ribbon is bent, the extent of the bending depends upon the vacuum, strength of the magnet, etc.



whereby a stream of rays could be caused, by means of a narrow slit in a mica or aluminum partition, to play upon a screen of some substance which would glow under their influence. The rays may be deviated by means of a magnet being attracted or repelled, according to the polarity. An electric field has the same influence; these rays are, therefore, different from light rays. Again, it may be demonstrated easily that these rays carry considerable energy with them. By using a concave cathode, the rays which are shot out at right angles to the surface, may be brought to a focus with the production of heat at that point. On making the current sufficiently strong, Sir William Crookes was able to fuse platinum-iridium by this means. Perrin calculated that the amount of energy produced by the impact of a kilogram of the corpuscles, assuming them to be particles, would raise to the boiling point in one second a lake 1,000 hectares (2,500 acres) in area and 5 metres deep.

Crookes's explanation of the phenomena recounted rests upon his assumption of material particles of residual gas in the tube, which being negatively electrified at the surface of the cathode are repelled by it and driven away with a high velocity. In support of the electrified radiant matter hypothesis, he devised a variety of tubes for demonstrating the mechanical action of the particles. Only one tube may be referred to here. It is known as the railway tube (Fig. 5). This tube contains a paddle wheel, whose axle rests upon a pair of glass rails. By making the proper terminal the



Fig. 5.

Fig. 5. Crookes' "railway tube." The cathode rays coming from one terminal drive the paddle wheel to the other end of the tube; by reversing the polarity, without disturbing the level of the tube, it is driven back.



cathode, the fly may be driven in either direction. Apparently in protest to the term "radiant matter," which implied the material nature of the particles, the Germans through the work of Goldstein, Wiedemann, and Lenard, invented the term, "cathode ray," which is in common use at present.

In 1895 Perrin<sup>1</sup> practically proved that the Crookes ray consists of streams of negatively electrified material particles. J. J. Thomson<sup>2</sup> confirmed these observations in 1897 and later actually measured their mass, electric charge, and the velocity of their movement. For full details one is referred to the original papers. For our purpose, suffice it to say, he<sup>3</sup> learned that the mass of each of the particles constituting the cathode rays is only one-thousandth the mass of the hydrogen atom, or  $2.3 \times 10^{-24}$  milligram\* and it has a velocity varying from one to four-tenths that of light,  $2.2-3.6 \times 10^9$  centimetres per second, depending upon the degree of exhaustion in the tube.

Thomson, further, showed that the ratio of the mass of a particle to its electric charge is independent of the nature of the gas and the electrodes. These particles, as a consequence, may be said to constitute a kind of matter previously unknown. The old Newtonian corpuscular theory being so strongly suggested, these particles have been called "corpuscles." They are constant constituents of all material atoms and molecules. Many chemists fail to coincide with the views of this eminent physicist, and few follow him to the extreme suggestion that these corpuscles constitute<sup>4</sup> negative electricity, which implies a return to the electric single fluid theory of Franklin.

In 1894 Lenard,<sup>5</sup> acting upon a suggestion of Hertz,<sup>6</sup> replaced a portion of the glass wall of the tube opposite the

1. Compt. Rend. 121, 1130.

2. Phil. Mag. V, 44, 293.

3. Phil. Mag. V, 547 (1899); Proc. Roy. Inst. 16, 574, (1901).

\*It has been calculated that the hydrogen atom weighs  $2.3 \times 10^{-21}$  milligrams.

4. Harper's Magazine 103, 564, Sept. 1901.

5. Ann. Phys. Chem. 51, 225; 56, 255, (1895).

6. Prof. Hertz observed that a very thin metallic film interposed inside a Crookes tubes permitted the glass to fluoresce under cathodic bombardment. The aluminum foil quite opaque to light, did not prevent this fluorescence of the glass behind it.

cathode with a very thin plate of aluminum (Fig. 6) and thus led the cathode rays out of doors, as it were, under ordinary pressure. If the Lenard rays are not a prolongation of the cathode rays, they are closely identified with them, for they can be deflected by a magnet, excite luminescence, and they affect a photographic plate. While the air is a turbid medium for them, they readily pass through thin sheets of aluminum, or even copper, and discharge an electroscope enclosed in a metal box. Lenard explored his rays by using a small luminescent paper screen covered with a wax-like organic chemical, pentadecylparatolyketone.

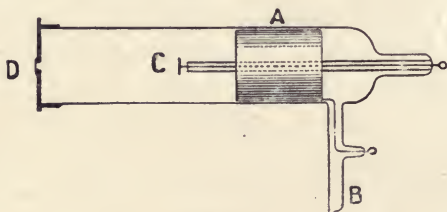


Fig. 6.

Lenard's tube. B is joined to a vacuum pump; A is anode; C cathode; D a thin aluminum window; From S. P. Thompson's "Light; Visible and Invisible." (Courtesy of the MacMillan Co.)

A year later Röntgen, while investigating cathode rays as studied by Hertz and Lenard, discovered that something came from his tube which caused a barium platino-cyanide screen lying on the table to luminesce strongly. Röntgen's tube had a greater vacuum, no window (Fig. 7) and was covered by a shield of black cardboard.<sup>1</sup> The cathode rays, cannot pass through glass. The X-rays, he learned, possess remarkable penetrative powers, readily passing through paper, wood, hard rubber, tin and aluminum foil. Silver, copper, platinum, gold and lead were less and less transparent to them, a plate of the last named 1.5 centimetres thick being quite opaque. Röntgen found that his rays affected a photographic

1. Uebereine neue Art von Strahlen (Vorläufige Mitteilung), Sitzungsberichte der Würzburger physik. medic. Gesellschaft, 1895. Nature 53. 274, (1896); Ann. Phys. Chem. 64, 1, 12, 18, 91, 898.

plate and that they differed from light rays, (visible, the ultra-violet, or infra-red, and any of those we have to consider), in not being reflected, refracted, or polarized.

Although Röntgen's investigation was very complete, there was one observation he failed to make, which was noted by several workers, shortly after his modest announcement, namely, that Röntgen rays, like the cathode rays, possess the

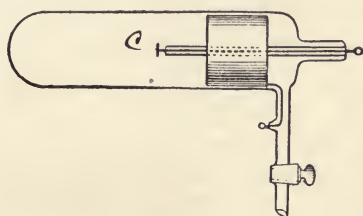


Fig. 7.

Röntgen's first tube. C. is the cathode. (Courtesy of the MacMillan Co.)

power of ionizing gases, when passed through them; that is, dissociating them into ions and rendering them electric conductors. A charged electroscope (Fig. 8) may be readily discharged by the surrounding air made a conductor by the passage of X-rays through it. Ordinary flame will also ionize air.

The effect upon a photographic plate gave a qualitative method for studying these rays, while the electrometric procedure presented a means of quantitative comparison.

If the tube be too highly evacuated, no current passes through at all, a vacuum being a perfect insulator. The explanation of the Röntgen rays at present accepted is that they are produced without the tube as a result of the bombardment of the cathode rays within. By the use of an anti-cathode, as shown in the modern Crookes tube, and by the automatic regulation of the gas pressure within, the effect of the X-rays may be accentuated (Fig. 9). A larger surface of the glass fluoresces and phosphoresces.

In an effort to learn the cause of the photographic effect

and especially considering if it might be attributed to the glowing of the walls of the tube, Henry<sup>1</sup> found that it could be

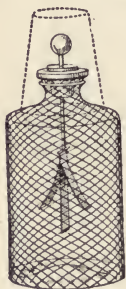


Fig. 8.

An electroscope is an instrument which illustrates that like kinds of electricity repel each other. The illustration shows thin strips of metal attached to a rod passing through a cork in a bottle. When a charged body is brought near the knob the leaves within are charged hence diverge. Anything which causes the air particles to become better conductors of electricity than ordinarily, will cause the leaves to collapse. The rendering of gases, as the air, a better conductor is known as ionization. Fig. 8 shows an Aluminum leaf electroscope, covered with a wire netting and metallic cap, which prevent its discharge by ordinary electric disturbances. The Röntgen rays discharge it whether charged by positive or negative electricity.

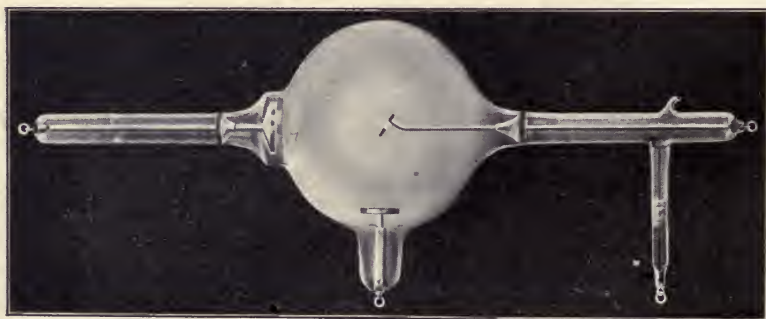


Fig. 9.

A modern Crookes tube for X-ray work. It is provided with a small side tube which contains a substance which absorbs or gives up sufficient gas to produce most penetrative effects.

1. Compt. Rend 122, 384, (1896).



augmented by phosphorescent zinc sulphide and Niewenglowski<sup>1</sup> observed that phosphorescing calcium sulphide would blacken a plate surrounded by light-tight paper. Troost<sup>2</sup> made a similar observation with Sidot's blende, and naturally occurring hexagonal zinc sulphide. H. Becquerel<sup>3</sup> (Figs. 10 and 11) found that calcium sulphide, the variety which phosphoresces blue-green to blue, would act strongly upon a photographic plate through two m.m. of aluminum foil, even though it were within a glass tube. W. Arnold<sup>4</sup> verified and extended these observations. The experiences of Madame Curie, Hofmann and Zerban, and ourselves did not accord with these observations.

As a result of a series of photographic experiments LeBon<sup>5</sup> concluded that sunlight generates in all bodies upon which it falls rays of "black light." The rays are invisible to the eyes. Their existence is shown by their action on a gelatinized silver bromide plate. Lumière, Becquerel and d'Arsonval<sup>6</sup> opposed this view and maintained that the black light is a



Fig. 10.

1. Compt. Rend. 122, 354, (1896).
2. Compt. Rend. 122, 564, (1896).
3. Comp. Rend. 122, 559, (1899).
4. Wied. Ann. 61, 316, (1897).
5. Comp. Rend. 122, 188, (1896).
6. Comp. Rend. 122, 500, (1896).

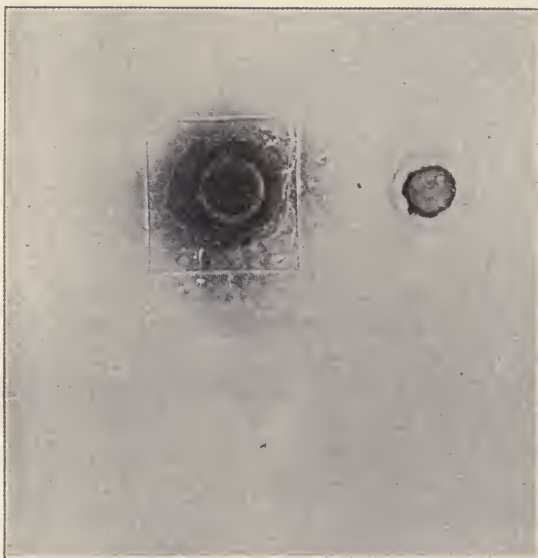


Fig. 11.

These two foregoing figures present small glass exhibition tubes containing various phosphorescing substances resting upon an aluminum shield, 2 m. m. thick, which is separated from the sensitive film of the plate by black paper. The second figure is the plate after forty-eight hours exposure. The "light" from the calcium sulphide and hexagonal blende did not penetrate the shields. (From Becquerel's paper).

kind of after light. They, also, put forward the idea that fluorescing substances, as for example the yellowish-green-glowing glass, are able to send out rays which penetrate dark bodies similarly to the Röntgen rays.

Physicists have long known that the salts of uranium luminesce most beautifully in the sunlight. Becquerel<sup>1</sup> next directed his attention to the double uranium potassium sulphate. This salt was placed on a plate so protected as to prevent the entrance of any sunlight. The whole was then

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<sup>1</sup>J. Compt. Rend. 122, pp. 420, 501, 559, 689, 762, 1086, (1896); 123, 835. Also address before the Roy. Inst., Great Britain, March 7, (1902).

exposed to the sun. On developing, the plate was found to be darkened. While preparing to repeat the experiment one day, it became cloudy. The whole apparatus was placed in a dark drawer where it remained during several days of inclement weather. For some unexplained reason the plate was developed without exposure to the sun at all. To his great surprise, he found that the plate had been distinctly affected. Becquerel's eminent father had shown years before that the phosphorescence of uranium salts persists but a very short time. Becquerel dissolved the double uranium potassium sulphate and purified it by recrystallization. The property of phosphorescence is not evident in solution. The entire process was carried out in the dark. He repeated the experiment with the photographic plate, being careful not to allow any exposure of the apparatus or material to light. (Figs. 12 and 13). Similar results to those noted above were obtained.

A number of uranium compounds were proved to possess this property (Fig. 14). The metal itself acted three and a half times as strong as the original sulphate. Further, while the light of the alkaline earth sulphides, zinc blende and other phosphorescing bodies gradually goes out in the dark, Becquerel proved that the property of uranium preparations of giving out rays, which penetrate light tight media, did not diminish even when they were kept for months in an absolutely dark place.

Becquerel's statements as to the "uranium rays" were almost immediately verified and extended by Spies,<sup>1</sup> Elster and Geitel,<sup>2</sup> Miethe,<sup>3</sup> Kelvin,<sup>4</sup> Beattie and de Smolan,<sup>5</sup> and Rutherford.<sup>6</sup> Later Hofmann and Strauss<sup>7</sup> and Crookes<sup>8</sup>, examined

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1. Verh. der physik. Ges. Berlin 15, 102, (1896).

2. Jahresber. Naturw. Braunschweig, 10, (1897).

3. Intern. fotogr. Monatsschrift f. Mediz. 4, 33, (1897).

4. Nature 55, 344, 447, (1896).

5. Phil. Mag. V, 43, 418 and 55, 277, (1897).

6. Phil. Mag. V, 44, 422, (1897) and 47, 109, (1899).

7. Ber. dtsh. Chem. Ges 33, 3126, (1900).

8. Proc. Roy. Soc. 66, 406, (1900).





Fig. 12.

Early Radiograph of an aluminum medal made by Becquerel with an uranium salt.



Fig. 13.

North Carolina uraninite (gummite) acted through a glass upon the plate in ninety hours.



Fig. 14.

Radiograph made by J. Collier of Denver with a pitchblende from the Wood mine of Leavenworth gulch, Gilpin county, Colorado. It is interesting as showing the relative transparency and opaqueness of different substances to the radium and uranium radiation. In this case the plate was not wrapped but enclosed in double light proof box, which was set in a dark room. Weight of pitchblende  $7\frac{1}{2}$  ounces; distance from plate, 4 inches; radiating surface,  $1\frac{3}{4}$  inches in diameter; exposure two weeks. Key to objects—(1) house finch; (2) imitation diamond; (3) real diamond; (4) cameo; (5) quartz crystal; (6) and (7) fluorspar; (8) Kauri gum; (9) tiger eye; (10) turquoise; (11) thick sheet lead; (12) thin sheet lead; (13) window glass; (14) centipede; (15) iceland spar; (16) amber; (17) black rubber. By courtesy of the Western Miner and Financier.)

many of the uranium minerals, (pitchblende, uraninite, bröggerite, cleveite, samarskite, and autunite, etc.), and found that they affected the photographic plate in the same manner. (Fig. 15).

It may be recalled that Röntgen rays are able to penetrate opaque sheets of metal, black paper, wood, caoutchouc, and so forth, and that they also have the property of ionizing gases.<sup>1</sup> The discovery showed that the Becquerel rays possessed the same properties. (Fig. 16).

1. Thomson and Rutherford, *Phil. Mag.* V, 42, 392, (1896).

It is well known that the components of sunlight are re-frangible and capable of polarization by means of tourmaline or Nicol's prisms. Rutherford<sup>3</sup> showed that the uranium rays possess this property in as limited a degree as the X-rays. This brilliant physicist, with Soddy,<sup>6</sup> showed the complex nature of the rays.

Those which are designated  $\beta$ -Rays have the following properties :

They are penetrative and affect the photographic plate.

They do not discharge the electroscope hence do not ionize gases and are but slightly absorbed by them.

When subjected to the influence of the magnetic field, they are bent like the cathode rays.

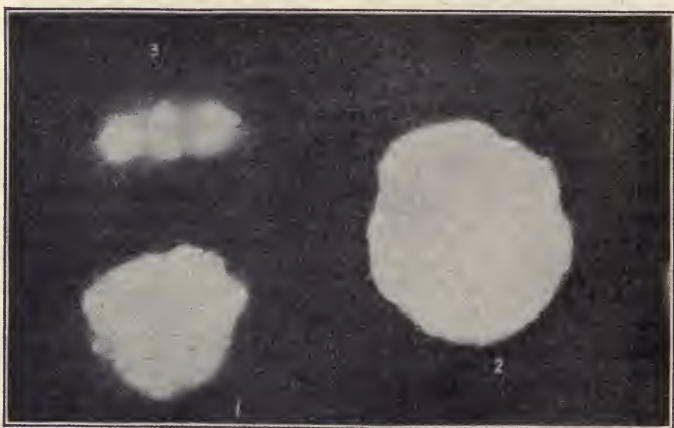


Fig. 15.

Carnotite impressions or flashes made by H. H. Buckwalter. Plate in double light-proof envelope. Time of exposure, one day. Nos. 1 and 2, carnotite concentrates, about seventy-five per cent. uranium from two per cent. ore. No. 3, very rich carnotite from the vicinity of Naturita, Colorado. (By courtesy of the Western Miner and Financier.

4. Compt. Rend. 124,800, (1897).

5. Phil. Mag. 47, 109, (1899).

6. Proc. Chem. Soc. 18, 121.



Fig. 16.

Radiograph made with Gilpin county (Colo.) pitchblende, by H. H. Buckwalter. Plate wrapped in two thicknesses of black paper. Time of exposure five days. About one-half pound of ore in two samples was used, separated from objects by a white pine board one inch thick. Dark circular object, an ordinary glass lens in chamois bag; square object, an aluminum box containing washers. Rays at greater angle passing through greater thickness cause apparent shadow. (By courtesy of the Western Miner and Financier.)

The <sup>ALPHA</sup> $\alpha$ -rays act thus:

They have no noticeable effect on the photographic plate.

They are responsible for most of the ionizing effect of the uranium preparations and are readily absorbed by different substances.

They are unaffected by the magnet.<sup>1</sup>

These facts, first established by Rutherford and Soddy, were subsequently recognized by Becquerel. (Fig. 17).

This phase of the subject will be reverted to in a later chapter and we shall put aside its further discussion until then.

1. This was later found to be incorrect, as will be shown.



For the present, let us assume the novel fact that energy comes continuously from uranium and its compounds. This energy loses nothing in intensity, even on keeping the radiant



Fig. 17.

The figure illustrates the absorption power of the different rays possessed by the screens of black paper, aluminum 0.10 m. m., and platinum 0.03 m. m. thick. The rays are deflected by a field about 1,740 C. G. S. units. The plate is unprotected except for the strips. The differences in penetration are readily noted. (After Becquerel.)

material in complete darkness for several years, as shown by Becquerel, and Elster and Geitel. To be sure as Rutherford has shown, this energy is small<sup>1</sup> and apparently spontaneous, unaffected by temperature,<sup>2</sup> and unchanged in liquid air ( $-180^{\circ}$  C). What is the cause of this unique physical phenomenon?

In 1897 Madame Sklodowska Curie of the École Municipale de Physique et de Chimie Industrielle at Paris, began an investigation of the relative activity of the various salts of uranium and later minerals bearing that element.<sup>3</sup> She measured the intensity of the radiation by its effect on the conductivity of the air unit. The apparatus is here described in her own words :

1. 1 g. of uranium oxide gives out in a year 0,032 Cal. of energy, Wied. Ann. Beibl. 24, 1,338, (1900).

2. Rutherford, Phil. Mag. 47, 109, (1899) and Becquerel, Compt. Rend. 130, 1,584 and 131, 137.

3. Compt. Rend. 126, 1, 101, April, 1898. See also her exquisite thesis presented to the Faculté des Sciences de Paris, which may be had in English for a small sum from the Chemical News of London, from which it has been reprinted.

“The method employed consists in measuring the conductivity acquired by air under the action of radio-active bodies; this method possesses the advantage of being rapid and of furnishing figures which are comparable. The apparatus employed by me for the purpose consists essentially of a plate condenser, A B (Fig. 18). The active body, finely powdered, is spread over the plate B, making the air between the plates a conductor. In order to measure the conductivity, the plate B is raised to a high potential by connecting it with one pole of a battery of small accumulators, P, of which the other pole is connected to earth. The plate A being maintained at the potential of the earth by the connection C D, an electric current is set up between the two plates. The potential of the plate A is recorded by an electrometer E. If the earth connection be broken at C, the plate A becomes charged, and this charge causes a deflection of the electrometer. The velocity of the deflection is proportional to the intensity of the current, and serves to measure the latter.

“But a preferable method of measuring is that of compensating the charge on plate A, so as to cause no deflection of the electrometer. The charges in question are extremely weak; they may be compensated by means of a quartz electric balance, Q, one sheath of which is connected to plate A and the other to earth. The quartz laminae are subjected to a known tension, produced by placing weights in a plate. The tension is produced progressively and has the effect of generating progressively a known quantity of electricity during the time observed. The operation can be so regulated that, at each instant there is compensation between the quantity of electricity that traverses the condenser and that of the opposite kind furnished by the quartz. In this way the quantity of electricity passing through the condenser for a given time, *i e.*, the *intensity of the current*, can be measured *in absolute units*. The measurement is independent of the sensitiveness of the electrometer.

“In carrying out a certain number of measurements of this kind, it is seen that radio-activity is a phenomenon capable of

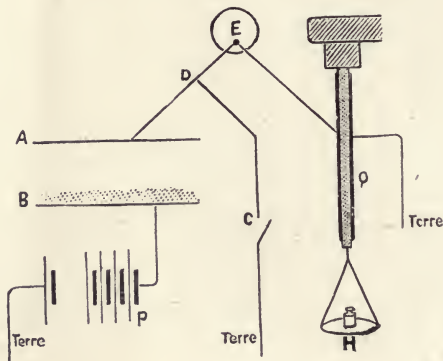


Fig. 18.

Plan of apparatus used by Mme. Curie for measuring the intensity of radiation from active bodies by their effect on the conductivity of the air. (From her thesis.)

being measured with a certain accuracy. It varies little with temperature; it is scarcely affected by variations in the temperature of the surroundings; it is not influenced by incandescence of the active substance. The intensity of the current which traverses the condenser increases with the surface of the plates. For a given condenser and a given substance the current increases with the difference of potential between the plates, with the pressure of the gas which fills the condenser, and with the distance of the plates, (provided this distance be not too great in comparison with the diameter). In every case, for great differences of potential the current attains a limiting value, which is practically constant. This is the *current of saturation*, or *limiting current*."

A discussion of the laws of conductivity of air and other gases subjected to the influences of the Röntgen and Becquerel rays, cannot be incorporated in a work of elementary character, so the reader is referred especially to the investigations of J. J. Thomson and Rutherford. The mechanics of the phenomenon appear to be the same in both cases and the theory agrees well with the observed facts. However, according to Townsend, the phenomenon is more complex when the pressure of the gas is low.



Using the term coined by the brilliant scientist, Madame Curie, the "radio-activity" of uranium and its compounds varied with the percentage of the metal present, as shown by Becquerel, hence the unique property was attributed to that element. Madame Curie verified the general conclusion, perfected methods of measurements and greatly extended the range of observation with consequent alterations. (Fig. 19). She measured all the common metals and non-metals, many rare compounds, and a large number of rocks and minerals. She found no simple substance other than uranium and thorium which gave evidence of atomic radio-activity. G. C. Schmidt<sup>1</sup> was the first to publish a statement as to the radio-activity of thorium. A striking fact is to be noted here. Thorium and uranium were the two elements then known to possess the highest atomic weights (232 and 240).<sup>2</sup> They frequently occur in the same minerals.

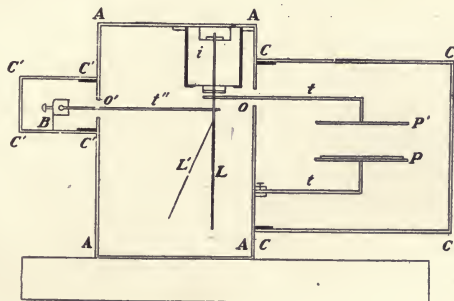


Fig. 19.

Electroscope used by Mme. Curie for qualitative examination of radio-active substances. The gold leaf  $L'$  when electrified through the terminal at  $B$  is repelled from the fixed metal strip  $L$ . Plate  $P$ , upon which the substance to be tested is placed, is connected with the metal case enclosing the apparatus. Plate  $P'$  is connected with the strips  $L$  and  $L'$ .

1. Wied. Ann. 65, 141, (1895).

2. It should be noted that white phosphorus, undergoing oxidation, according to Black, causes the air to become a conductor. As neither the red variety nor compounds of phosphorus exhibit this property, it is readily attributed to chemical action and cannot come into consideration here.

When the air between the plates becomes ionized a charge passes across and  $I$  falls. By rating the time necessary for collapsing, or by having a scale at the back and rating the time required for the leaf to pass through a selected number of divisions, the radio-activity may be approximately determined, *e. g.*, that portion which ionizes gases. There are several forms of apparatus using the same principle. The limits of this book will not admit of their description, other than to call attention to Rutherford's variation, namely, the leaves are insulated from the rest of the apparatus being suspended by means of a sulphur bead. After charging through the conductor  $'''$ , it is swung aside. The rate of leakage is thus reduced to a minimum.

The following is a table made by Madame Curie giving in  $10^{-11}$  amperes the intensity of the current obtained with metallic uranium and with different minerals.

	<i>i</i>
Uranium . . . . .	2.3
Pitchblende from Johanngeorgenstadt . . . . .	8.3
Pitchblende from Joachimsthal . . . . .	7.0
Pitchblende from Pzibram . . . . .	6.5
Pitchblende from Cornwallis . . . . .	1.6
Cleveite . . . . .	1.4
Autunite . . . . .	2.7
Chalcolite . . . . .	5.2
	0.1
	0.7
Various thorites . . . . .	1.3
	1.4
Orangite . . . . .	2.0
Monazite . . . . .	0.5
Xenotime . . . . .	0.03
Aeschnite . . . . .	0.7
	0.4
Fergusonite (two samples) . . . . .	0.1
Samarskite . . . . .	1.1
Niobite (two samples) . . . . .	0.3
Tantalite . . . . .	0.02
Carnotite . . . . .	6.2

All the minerals having radio-activity contained uranium and thorium. A glance at the table, however, shows the amazing fact *that certain minerals possess a greater intensity than the metal uranium itself.* This is utterly at variance with what we have already learned, namely, that the radio-

activity is dependent upon the percentage of the metal, uranium or thorium, present.

Afanasjew<sup>1</sup> examined fifty-one minerals by their action on a photographic plate. All the minerals containing uranium and thorium blackened the plate. Pisani<sup>2</sup> made somewhat similar experiments and raised the question,—Is this astonishing state of affairs due to the small percentage of the oxides of uranium and thorium, or is it caused by the presence of a new radio-active body?

To throw light on the subject, Madame Curie prepared artificial chalcocite, a double copper uranium phosphate, from pure materials. It showed normal activity, namely two-and-a-half less than uranium, instead of 5.2 as great. Pisani's question was answered. Madame Curie's inevitable conclusion was that pitchblende, chalcocite and autunite contained a small quantity of a strongly radio-active body, differing from uranium and thorium, differing from any of the elementary bodies known.

The difficult problem which confronted this intrepid woman was the seeking of a new element, each faltering step being guided by a veritable fairy wand. The glorious outcome of her researches joined the century most replete with human achievement to another, which promises even more and greater marvels.

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1. J. Russ. Phys. Chem. Soc. 32, ii, 103 (1900).

2. Bull. Soc. franc. Mineral, 27, 58.

## CHAPTER II.

## THE EXTRACTION OF RADIUM; ITS PROPERTIES, PHYSICAL AND CHEMICAL.

Pitchblende is an expensive mineral mined mainly in Bohemia for the uranium it contains. It is one of the most complex ores; containing besides the uranium, iron, calcium, lead, aluminum, silicon, copper, bismuth, zinc, cobalt, nickel, manganese, antimony, arsenic, vanadium, thallium, columbium, tantalum, many rare earths, and so forth. In the course of her search for the cause of the unique properties possessed by the uranium bearing minerals, Madame Curie<sup>1</sup> obtained a very radio-active substance, resembling bismuth, which she named, polonium, after her native country. This will be dealt with later. For this work she received the Gegner prize of 4,000 francs. At this point her husband having joined in the work, they were assisted by M. Bemont, Director of the Ecole Municipale.<sup>2</sup> Pitchblende, in sufficient amount, being beyond the purse of the teacher she secured from the Austrian Govern-

1. Compt. Rend. 127, 175. Rapports au Congrès International de Physique, III, 79, Paris (1900).

2. Madame Sklodowska Curie was born in 1867, at Warsaw, Poland, where she received her early training. In 1891 she went to Paris, continued her studies at the University and received her "Master's" degree in Physics and Mathematics. She married Professor Pierre Curie, the Professor of Physics at the University of Paris, in 1895. The year following she successfully qualified as a candidate for a professorship in a girl's college. In 1900 she was appointed Professor of Physics in the State Normal School for Women at Sèvres. She received her "Doctor's" degree recently for the thesis already quoted. The writer knows of no doctorate dissertation of such scope, elegance, breadth of conception and importance in its contribution to knowledge.



ment a ton of the "tailings" or residues of the ores from which the uranium had been extracted.

To extract the uranium, the process is as follows: The crude ore is crushed, roasted with sodium carbonate, washed with warm water and then with sulphuric acid. The solution contains the uranium. The insoluble residue, "tailings," which is rejected, contains most of the bodies of high radio-activity; its activity being four and a-half times that of metallic uranium. Laboratory methods not being easily applied, M. Debierne organized the treatment in the factory, which was erected at Ivry without the walls of the city of Paris.

A ton of the residues was worked up and a few decigrams of a substance resembling barium obtained. This was many thousand times as active as uranium. Although this substance showed but slight change in its atomic weight from that of barium, and no characteristic new lines were to be seen in the visible spectrum, according to Demarçay, it glowed feebly in the dark, affected a photographic plate through black paper and even thin sheets of metal, and ionized gases. These facts were sufficient to warrant the assumption of the presence of a new element. It was named radium.<sup>1</sup>

So far, although Phillips,<sup>2</sup> Boltwood,<sup>3</sup> and others have proposed methods for analyzing ores for radium, it has not been extracted commercially in America. Lockwood, at Buffalo, has installed a plant for its extraction from carnotite, but as yet no preparations from that factory have been placed upon the market.

The importance of the discovery of radium, the minute percentage in which it is found, the extremely unique properties possessed by its compounds, the desire for it on the part of experimenters and even the merely curious, and its possible utility in medicine have created a radio-mania. The demand

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1. Compt. Rend. 127, 1215.

2. American Phil. Academy, April Meeting, (1904). Science, May 6th.

3. Eng. Min. J. May 12, (1904).

Boltwood (Eng. & Min. Jour. 77, 756) tested for the presence of radium in uranium compounds as follows:

has been far in excess of the supply. The prices have almost tripled. The Austrian Government has forbidden the shipping of uranium ores or tailings without that country. Many tons of uranium ores have been shipped from the United States to Europe. The United States Geological Survey in seeking the locations of all the uranium bearing ores, has with its usual progressiveness appointed a special expert<sup>1</sup> and issued a letter on the subject. It says :

"The simplest means of detecting radio-activity in a substance is by use of the photographic plate. The more sensitive the plate the better. The plate should not be removed from the enclosing black paper, and a metal object should be laid upon this black paper in a dark room; upon this should be placed the specimen to be tested. Instead of the metal object a few small nails may be arranged so as

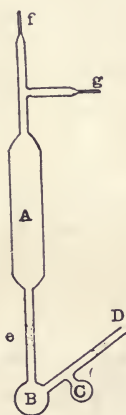


Fig. 21

"A piece of apparatus (Fig. 21) constructed entirely of glass was first prepared. This consisted of a bulb (A) of about 50 cubic centimeters capacity, which was joined by a short tube to a smaller bulb (B). An accurately weighed quantity of the very finely powdered mineral was introduced into the bulb B, and in the bulb C was placed a sufficient quantity of a suitable acid, its actual quantity and nature depending on the character of the particular mineral under investigation. The whole apparatus was then sealed up air-tight at a slightly diminished pressure and, by tilting, the acid was transferred to the bulb B, containing the mineral. The mineral was then completely decomposed by gentle warming and the apparatus was allowed to stand for several days to permit the radium emanation, which is freed when the radium salts pass into solution, to diffuse uniformly through the interior of the apparatus. The bulb A was then sealed off from the rest of the apparatus, allowed to stand for two hours, in order that any rapidly decaying emanations (actinium and thorium) which it contained might completely decompose, and, after washing the interior walls with a strong sodium hydroxide solution to completely remove acid fumes, the air and radium emanation which it contained was transferred to an air-tight electroscope, and the rate of the leak measured."

The sensitiveness of the method is extraordinary. Dr. Boltwood was able to compare the relative quantities of radium in two samples of pitchblende weighing from .001 to .002 gram. He was able to detect the presence of .000,000,000,1 gram of radium. It is probably possible to quantitatively estimate the quantity of radium equal to perhaps 1-100th of the above.

1. Dr. Geo. F. Kunz, 40 East 25th St., New York City.



Fig. 21a.

Radium Exhibit of U. S. Geological Survey, Dr. G. F. Kunz, Special Agent, Louisiana Purchase Exposition, St. Louis, Mo., U. S. A. (By courtesy of the Survey.)

to form the initial of the owner and left on the paper-covered plate below the specimen. The specimen should be left in the dark room from two to fifteen hours and then developed in the usual manner. If the specimen has radio-active powers, a photograph of the metal object or of the nail-formed initial will be produced on the plate exactly as if it had been exposed to the sun's rays. The test should be made, if possible, with from half a pound to a pound of the material. The electrical method is more reliable, but is much more difficult."

M. Jacques Danne,<sup>1</sup> Preparator for Mme. Curie, says: "The extraction of the radium salts from pitchblende and carnotite takes place in three stages. The first stage consists in the roasting of the uranium ores, the preliminary roasting without soda, the final roasting with soda and a little saltpetre. The ores are then treated with sulphuric and a little nitric acid, and the resulting solution contains the uranium salts, while the radio-active metals are contained in finely divided form in the residue as sulphates. The residue is then treated with concentrated hydrochloric acid and a part of it goes into solution. This solution contains the greater part of the elements polonium and actinium. Polonium is precipitated with sulphuretted hydrogen and in the filtrate the actinium is thrown down with ammonia after oxidation. The residue from the treatment with hydrochloric acid, which contains the radium, is washed with water and treated with concentrated boiling soda solution, in order to change the sulphates, which were left undecomposed by the treatment with hydrochloric acid, into carbonates. The residue is washed with water and digested with pure hydrochloric acid. The solution resulting from this treatment contains radium and a little polonium and actinium. After filtration the solution is treated with sulphuric acid, which throws down a mixture of sulphates of radio-active barium, lead, calcium, and a little actinium. One ton of uranium residues furnished about 10 to 20 kilograms of the mixtures of sulphates, the radio-activity of which is 30 to 60 times greater than that of metallic uranium. The mixture of sulphates is treated with boiling, concentrated soda solution, and the carbonates

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1. Genie Civil, Jan. 16 (1904).



thus obtained are converted into chlorides by hydrochloric acid. Sulphuretted hydrogen is introduced into the solution, whereby a small precipitate of active sulphides which still contain the polonium is formed. The filtrate from this precipitate is then oxidized with potassium chlorate and precipitated with ammonia; the hydrates and oxides thus precipitated still contain actinium. The filtrate is treated with soda to precipitate the carbonates of the alkaline earths, this precipitate being then converted into chlorides and the solution evaporated to dryness. The dry residue is treated with concentrated hydrochloric acid, when the radio-active barium chloride and the radium chloride remain insoluble. This residue of chlorides is then dissolved in water, the carbonates precipitated by adding soda, and the precipitate treated with hydrobromic acid in order to convert the carbonates into bromides. A ton of material treated in this way furnishes about 8 to 10 kilograms of radio-active barium bromide nearly 60 times more radio-active than metallic uranium. The bromide is now subjected to a long series of crystallizations,<sup>1</sup> dependent upon the fact that the radium bromide is less soluble in water than the barium bromide. Each crystallization furnishes a product which has a greater radio-activity than the preceding and this treatment is kept up until the desired radio-activity is reached."

### PHYSICAL PROPERTIES OF RADIUM SALTS.

The element radium has not yet been obtained. Wedekind<sup>2</sup> and Marckwald<sup>3</sup> have, however, prepared radium amalgams; the former by electrolysing a halogen compound in the presence of mercury, and the latter by treatment with sodium amalgam. No description of the metal can be given and we do not know certain of its properties, as the specific gravity, melting point, etc. Curie says it is a mere matter of obtaining sufficient of the chloride, when it may be prepared like the alkaline earth

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1. As first suggested and done by Giesel.

2. Chem. Zeit. 28, 269 (1904).

3. Berichte, Chem. News, 89, 97.

metals. The difficulty of working with large quantities are very apparent when we realize that only a few centigrams of fairly pure material are had from two tons of the ore, and are aware of certain properties soon to be mentioned.

The chloride, bromide, carbonate, acetate, nitrate, and sulphate of radium freshly prepared resemble similar salts of barium. The nitrates of radium and of barium are about equally soluble in water. The halides are isomorphous, but differ slightly in their solubility in water.

Radium salts gradually assume color. Apparently they undergo alterations through the influence of the rays they emit, giving out oxygenated chlorine compounds, if the salt be a chloride. Giesel has shown that a water solution of a radium salt gives off hydrogen continuously.

The earlier prepared compounds of radium were much contaminated with barium and gave an atomic weight of 137.5, barium being 137.35. Successive fractionations gave 146,<sup>1</sup> 175,<sup>2</sup> and finally 225.<sup>3</sup>

In order that a new substance may claim a place in the family of chemical elements, it has been agreed that it must give a characteristic spark spectrum. A preparation not very strong was submitted to Demarcay,<sup>4</sup> who found in addition to the barium lines, a new one in the ultra-violet. With purer materials that lamented chemist photographed a characteristic spectrum, which is in general similar to the alkaline earths. With radium bromide prepared by Giesel, Runge<sup>5</sup> and Precht, Exner and Haschek<sup>6</sup> obtained the spark and flame spectra of the characteristic carmine-red coloration given by the Bunsen flame. Their work was concerned largely with the visible

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1. Compt. Rend. 129, 20.

2. Compt. Rend. 131, 6.

3. Compt. Rend. 135, 161 (1902).

4. Compt. Rend. 127, 1218 (1898); 129, 716 (1899); 131, 258 (1900).

5. Astrophys. Journ. 1 (1900).

6. Sitz. Ak. Wiss. Wien. July (1901).

spectrum, while Demarçay observed the ultra-violet in the main. Recently Crookes<sup>1</sup> made an elaborate and extended study of the spark spectrum in the ultra-violet region. Runge and Precht<sup>2</sup> noted the influence of a magnetic field on the spectrum and that it was composed of series analogous to calcium, strontium, and barium. As these series appeared to be connected with the atomic weights, they calculated that the atomic weight of radium should be 258. By utilizing the relation between the spectra of some elements, and the squares of their atomic weights, Watts<sup>3</sup> arrived at the same value given by Mme. Curie. Although Runge and Precht<sup>4</sup> have criticized, perhaps justly, the method used by Marshall Watts, the value 225 is accepted for several reasons, one being that that value causes it to fall in the family of alkaline earths in the periodic system of Mendeleeff.

A radium compound, within a closed glass tube, when brought near a screen of zinc sulphide, or barium-platino-cyanide, causes it to glow brightly in a dark room. Photographic plates, covered with black paper, are at once affected.



Fig. 22.

Radium bromide within a closed glass tube affects the photographic plate through black paper. Pacini in our laboratory used the radium tubes as a pencil and traced the above.

1. Sitz. Kgl. Pr. Akad. Wiss Berlin (1904), 417.
2. Phil. Mag. April (1903).
3. Phil. Mag. IV, 5, 203 (1903).
4. Phil. Mag. IV, 5, 476 (1903).

(Fig. 22.) All the radium compounds, so far obtained, are luminous in the dark. We do not know whether radium itself actually gives out luminous rays or whether the luminosity results from the conversion by the solid substance itself, or the impurities present, of invisible rays into those which give the effect of light on the optical organs. The presence of radium causes certain substances, as Thuringian glass, diamonds, willemite, kunzite, etc., to fluoresce and phosphoresce.

The salts of radium appear to be a source of spontaneous and continuous evolution of heat. Curie and Laborde<sup>1</sup> first showed that the temperature of an impure radium salt is  $1.5^{\circ}$  C.

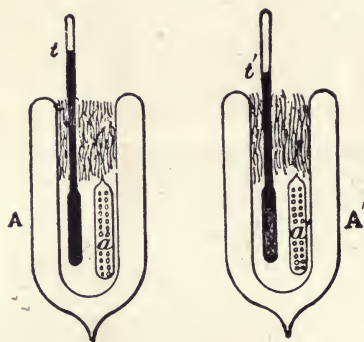


Fig. 23.

Simple method for illustrating the continuous disengagement of heat by radium. Delicate thermometers ( $t$  and  $t'$ ) in duplicate are placed within calorifically isolated vessels (Dewar bulbs),  $A$  and  $A'$ . Small tubes of equal size,  $a$  and  $a'$ , containing molecular weights of, say, radium and barium chlorides, the latter being inactive, are inserted after thermic equilibrium has been established. (After Curie, see Danne.)

1. Compt. Rend. 136, 673 (1903).



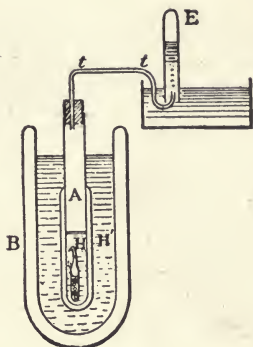


Fig. 24.

The quantity of heat given out by a radium salt has been determined by the apparatus of Dewar and Curie, shown above. A small thin-walled Dewar bulb, A, containing liquid hydrogen, is immersed in liquid hydrogen, H', within a larger thermic insulator, B. Tube H by means of glass tubing ends underneath the eudiometer, E, over water. No gas escapes through the exit tube, t, until the tiny glass vessel containing the radium compound is inserted, after which there is a continuous and regular ebullition. 0.7 gram of radium bromide causes 70 c.c. of hydrogen to be evolved every minute. Freshly prepared salts disengage relatively smaller amounts of heat.

higher than the surrounding medium (Fig. 23). Later Curie<sup>1</sup> found 3° and Giesel 5° C. difference for the bromide. The former, also, learned that the rate of the emission of heat depended upon the age of the compound. When the compound is freshly prepared the emission is small. It increases and reaches a constant maximum in a month. He also learned that if the salt be dissolved in water and placed in a sealed tube, that the difference in condition made no difference in the emission of heat. By the use of a Bunsen calorimeter, or the other method shown in the illustration (Fig. 24), Curie and Laborde learned that one gram of a pure radium salt emits about 100 gram-calories of heat per hour. Runge and Precht<sup>2</sup> with others

1. Societe de Physique. (1903).

2. Sitz. Ak. Wiss. Berlin, (1903).



Fig. 25.

Skiagraph of tools made with radium bromide, 300,000 activity. Eight inches separated the plate and tube. Exposure forty minutes.

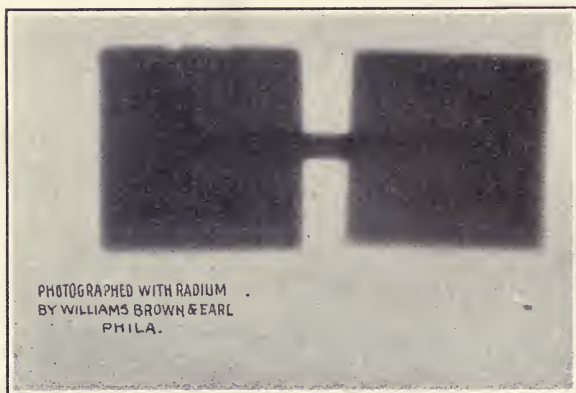


Fig. 26.

Showing penetration of radium rays. A lead bar was placed beneath plates of cast iron  $\frac{1}{2}$  inch thick (Brown).

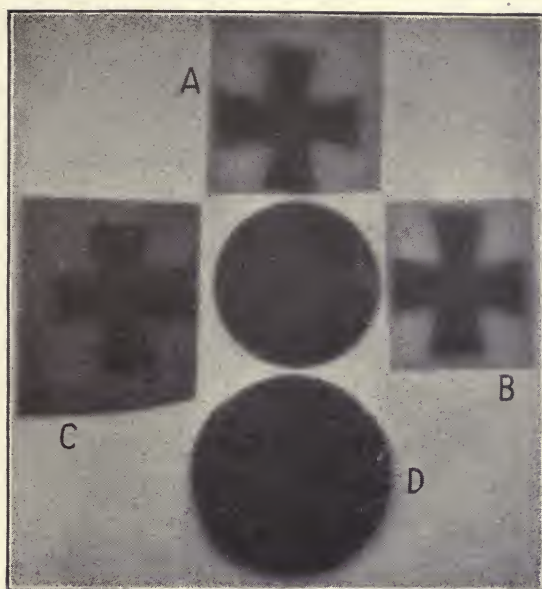


Fig. 27.

Illustrating the penetrability to radiations of radium of Aluminum (A), Micro-cover (B), Micro-slide (C), Red flash-glass (D), and a Silver Quarter (in the center).

confirmed this continuous emission of heat. One gram of radium emits in a day 2400, or in a year 876,000 gram-calories.

The radiations given out in part penetrate paper, thin metals, thick metals, glass, mica, etc. This wonderful phenomenon has been studied by Strutt,<sup>1</sup> the Curies,<sup>2</sup> and others. Most striking experiments, illustrating the penetration of the rays, are easily performed. (Figs. 25, 26 and 27.) Hammer<sup>3</sup> placed a tube of 7000 activity within a cannon ball, sealed it, and

1. Nature, 39, (1900).

2. Loc., Cit.

3. Radium and other Radio-active Substances, Lecture before the S. E. E. & Am. El. Ch. Soc., April (1903).

made a skiagraph. Kunz and the writer<sup>1</sup> caused a large tiffany-ite diamond to glow when radium bromide (300,000 activity) was protected by covers of glass, gutta-percha, steel tubing, three sheets of copper, one m.m. of silver and ten c.m. of water.

Radium compounds are the first chemical preparations known to spontaneously charge themselves with electricity. Placed near electrically charged and isolated bodies, as an electroscope, they discharge them. (Fig. 28.) Thus they ionize gases, which property serves as a most delicate means of determining in part the activity quantitatively.<sup>2</sup> (Fig. 29.)

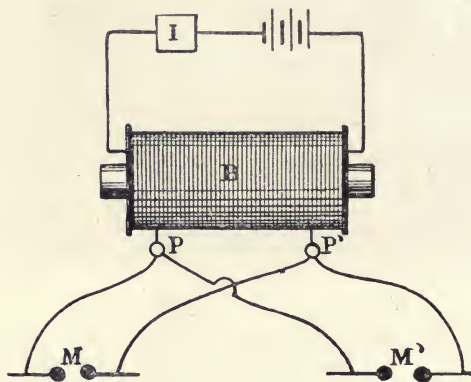


Fig. 28.

This figure illustrates a beautiful experiment of Professor Curie's, which shows the conductivity of the air under the influence of radium. The secondary terminals, P P', of an induction coil, B, are connected by wires with two sets of electrodes, M and M', so separated as to offer two paths for discharging sparks. If a tube of radium be brought near one set, while the sparks are passing rapidly between both pairs, the sparks will cease at the second set as the path offered, where the radium is present, is much less resistant than the normal air at the other.

1. Science. N. S. 18, 769, (1903).

2. The term "ionization," as here used, has no reference to the modern theory of solutions, but to the interpretation given by J. J. Thomson.



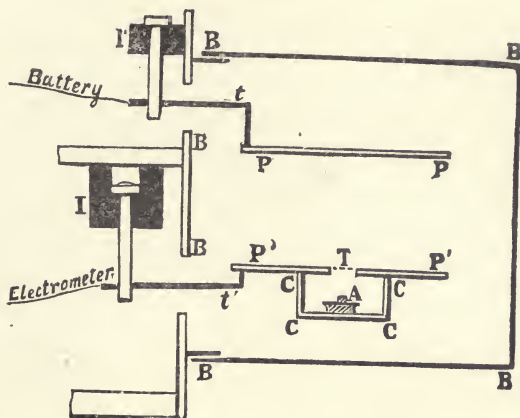


Fig. 29.

The apparatus used by the Curies for the determination of electrical conductivity is described in the words Madame Curie as follows :

“The two plates of a condenser, P P and P' P' (Fig. 29), are horizontally disposed in a metallic box, B B B B, connected to earth. The active body, A, placed in a thick metallic box, C C C C, connected with the plate P' P', acts upon the air of the condenser across a metallic sheet, T; the rays which pass through the sheet are alone utilized for producing the current, the electric field being limited by the sheet. The distance, A T, of the active body from the sheet may be varied. The field between the plates is established by means of a battery. By placing in A upon the active body different screens, and by adjusting the distance A T; the absorption of rays which travel long or short distances in the air may be determined.”

Attention must be directed at this point to an interesting phase of the investigation of these radio-active bodies. Coppel<sup>1</sup> has determined that by means of the spark spectrum one may detect 1 part in 900,000 of barium and 1 in 100,000,000 of strontium. The principal line of radium in the ultra-violet may be seen faintly in a preparation 40 times as active as uranium. By the electrical method, depending upon the ionization of the air, the presence of radium in a sub-

1. Pogg. Ann. 628, (1870).

stance may be detected when it possesses only 1/1000 the activity of uranium. With the most sensitive electrometer 1/10,000 the activity of uranium may be observed. Thus we see that radio-activity is a detectable property nearly a million times more sensitive than spectrum analysis, which is at least a thousand times more sensitive than the most delicate balance. This should not excite great surprise. Berthelot<sup>1</sup> has called attention to a comparison of the delicacy of detecting radio-activity and odors. 1/100 billionth of a gram of iodoform is readily detected by a sensitive nose.

Crookes<sup>2</sup> separated from uranium and the writer from thorium<sup>3</sup> a fraction which did not affect the photographic plate. Rutherford<sup>4</sup> showed that this portion, which did not affect the sensitive gelatine, continued to ionize gases. In short, the radiations were proved to be complex. (Fig. 30.)

As a result of numerous investigations, by different workers, but mainly Rutherford, the radiations from radium have been found to consist of three types of rays :

1. Those which are easily absorbed ( $\alpha$ -rays) ;
2. Those which are penetrating ( $\beta$ -rays) ;
3. Those which are very penetrating ( $\gamma$ -rays).

Rutherford<sup>5</sup> found, both in uranium and thorium, rays which differed in their penetrating powers. He designated them  $\alpha$ - and  $\beta$ - rays. Later the very penetrating rays were obtained from these two elements and radium and designated  $\gamma$ -rays. The term "ray" is applied to a stream of corpuscles, such as Newton pictures in his theory of light.

The  $\alpha$ -rays correspond to the canal rays of Goldstein which, according to Wien, consist of positively charged particles, projected with great velocity. The  $\beta$ -rays are the same as the cathode rays, while the  $\gamma$ -rays, in some respects, resem-

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1. Compt Rend. 138, 1249.
  2. Proc. Roy. Soc., 66, 409.
  3. J. Am. Chem. Soc. 23, 761. (1901).
  4. Phila. Mag. (1901).
  5. Phil. Mag., Jan. (1899).

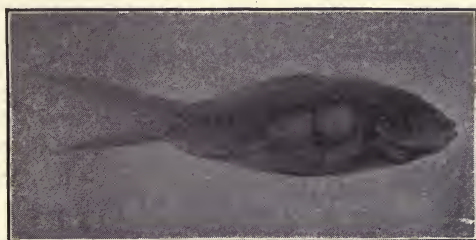


Fig. 30.

Radiograph of a fish obtained by an exposure of 40 minutes with radium of 300,000 activity. The lack of definition is noticeable, as the  $\beta$ -rays were not separated. The lower of the two pictures is a radiograph of the same fish made by the Roentgen rays.

ble the Röntgen rays. The Röntgen rays result from the expenditure of electric energy within a vacuum tube. They vary with the conditions, whereas those given out by radio-active bodies are apparently emitted spontaneously and at a rate not influenced by any chemical or physical agencies. The velocity and penetrating powers of the rays from radio-active bodies appear to be greater than those produced in a vacuum tube. The method used by Madame Curie for illustrating these rays is shown in Fig. 32.

The ionization effect of the  $\alpha$ -,  $\beta$ - and  $\gamma$ -rays is in the order 10,000 : 100 : 1. The penetrating power of the rays

is as follows: A sheet of aluminum 0.0005 c.m. thick will cut off one-half of the  $\alpha$ -rays: 0.05 c. m., one-half of the  $\beta$ -rays, and 8 c. m., one-half of the  $\gamma$ -rays; or, in short, it will be noted that ionization and penetration powers bear an approximately inverse ratio. The making of comparative measurements is fraught with numerous difficulties, so the figures are only approximate.

For reasons that will become apparent, these rays will be considered in the order of their conduct under the influence of a magnetic or electro-magnetic field.<sup>1</sup>

#### THE $\beta$ - OR CATHODE RAYS.

Elster and Geitel observed that the conductivity produced in the air by radium rays was affected by a magnetic field. Giesel<sup>2</sup> demonstrated that the rays deviate under the influence of an electro-magnet in the same direction and in the same order of magnitude as the cathode rays. Meyer and von Schweidler<sup>3</sup> verified this later and Becquerel,<sup>4</sup> using the photographic method, demonstrated the magnetic deflection of the rays. (Fig. 31.) Rutherford<sup>5</sup> demonstrated that the rays from uranium consisted of  $\alpha$ - and  $\beta$ -rays.

P. Curie, by the electrical method, showed that radium rays consisted of non-deviable and easily absorbed ( $\alpha$ -rays) and penetrating, but deviable by the magnetic field, ( $\beta$ -rays). Rutherford and Grier, also using the electrical method, demonstrated that thorium compounds gave in addition to the  $\alpha$ -rays some penetrating  $\beta$ -rays, deviable in the magnetic field as in the case of uranium. The ionization produced by the  $\alpha$ -rays is large in comparison to that due to the  $\beta$ -rays.

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1. For a complete discussion of the methods of measuring ionization of gases the reader is referred to special works like J. J. Thomson's "Conduction of Electricity Through Gases," and Rutherford's "Radio-activity."

2. Wied. Annal. 69, 831, (1899).

3. Phys. Zeit. I, 90, 113, (1899).

4. C. R. 129, 997, 1205, (1899).

5. Phil. Mag., Jan. (1899).

6. Phil. Mag., Sept. (1902).



Fig. 31.

The radium preparation is placed in a small lead cup open above. The rays are projected like the smoke from a mortar. Under the influence of a powerful magnet or electro-magnet a portion are attracted, a portion repelled, and the remainder, being unaffected, continue in a straight line. (After Curie.) See next figure for a more graphic illustration and the nomenclature of the rays.

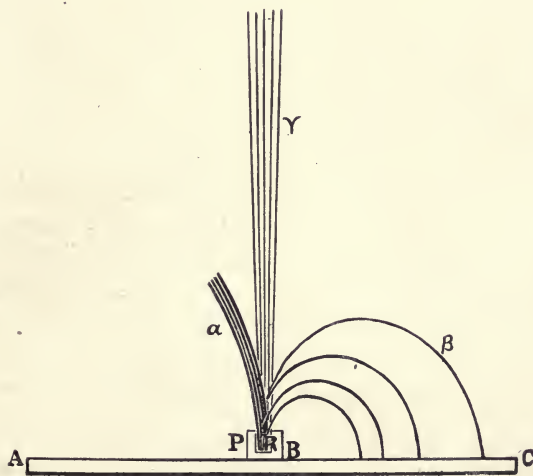


Fig. 32.

A graphic illustration of the radiations of radium. The active preparation (R) is in a lead cup. See description in the text.



THE  $\alpha$ -RAYS.

The ease with which the  $\beta$ -rays were deviated by the magnetic, or electro-magnetic field and their penetration naturally commanded the greater attention at first. A magnetic field strong enough to produce a marked deviation of the  $\beta$ -rays, had little or no effect upon the  $\alpha$ -rays. In fact they were regarded as secondary rays, set up by the  $\beta$ -rays in the active matter from which they were produced. It was learned, as adverted to, that the matter giving rise to the  $\beta$ -rays could be separated from uranium, while the intensity of the  $\alpha$ -rays was not affected. Strutt<sup>1</sup> and later Crookes<sup>2</sup> suggested that the  $\alpha$ -rays might consist of positively charged bodies projected with great velocity. Madame Curie,<sup>3</sup> from her study of polonium, suggested the probability that these rays were bodies moving very rapidly, but losing their energy when they passed through matter. Rutherford<sup>4</sup> learned, by most careful experimentation with the electrical method, that the  $\alpha$ -rays could be deflected by an intense magnetic field and in the opposite direction from the cathode rays, and demonstrated that they consisted of positively charged particles.

Becquerel<sup>5</sup> confirmed this by the photographic method. The naked radium preparation was covered with a metallic screen over a narrow slit. The photographic plate was placed two c.m. above this slit. The strength of the magnetic field was great enough to deflect all the  $\beta$ -rays. The plate was affected not only immediately opposite the slit, but also on the side away from the magnetic field. On reversing the field for equal lengths of time the image, which had been produced by the  $\alpha$ -rays, was observed to be reversed also. Descoudres<sup>6</sup>

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1. Phil. Trans. 507, (1901).
  2. Chem. News 85, 109, (1902).
  3. C. R. 130, 76, (1900).
  4. Phys. Zeit. 4, 235, (1902).
  5. C. R. 136, 199, (1903).
  6. Phys. Zeit. 4, 483, (1903).



proved that the  $\alpha$ -rays of polonium are deviated in the same manner.

Wien has shown that the velocity of the projection of the canal rays varies with the gas in the tube and the intensity of the electric field applied. It is generally about one-tenth of the velocity of the  $\alpha$ -rays from radium. For the  $\alpha$ -rays of radium it has been shown that  $v = 2.5 \times 10^9$  and  $e/m = 6 \times 10^3$ ,  $10^4$  is the value of  $e/m$  for the hydrogen atom liberated in the electrolysis of water. If the charge of the  $\alpha$  particle be the same as that of the hydrogen atom, the mass of the  $\alpha$  particle is about twice as great as that of the hydrogen, which would indicate that it consists of either helium or hydrogen. This phase of the subject will be taken up in the fourth chapter.

The  $\alpha$ -rays, coming from different sources, vary in the amount of their absorption. About ninety-nine per cent. of the ionization of the air produced by naked radium is due to the  $\alpha$ -rays<sup>1</sup>. The order of the penetration of the  $\alpha$ -emanations as found is: thorium and radium (excited radiation), thorium, radium, polonium and uranium. The substances used were: aluminum, Dutch metal, paper; air and other gases.

### $\gamma$ - OR VERY PENETRATING RAYS.

Villard<sup>2</sup> and subsequently Becquerel<sup>3</sup> discovered by using the photographic method these very penetrating rays, which are non-deviable by a magnetic field. Rutherford,<sup>4</sup> by using the electroscope of C. T. R. Wilson,<sup>5</sup> found that uranium and thorium also gave out  $\gamma$ -rays.

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1. Rutherford, Phil. Mag., Jan. (1899).  
Owens, Phil. Mag. Oct. (1899).  
Rutherford and Brookes, Phil. Mag. July, (1902).
  2. C. R. 130, 1110, 1178, (1900).
  3. C. R. 130, 1154, (1900).
  4. Phys. Zeit. pp. 517, (1902).
  5. Proc. Roy. Soc. 68, 152, (1901).

As a result of the investigations of Benoist,<sup>1</sup> Strutt,<sup>2</sup> and others, it is known that the  $\gamma$ -rays possess great penetrating power; that they are non-deviable in an intense magnetic field; that  $\gamma$ -rays and  $\beta$ -rays occur together and in the same proportion; that they seem to be absorbed in a similar way to the

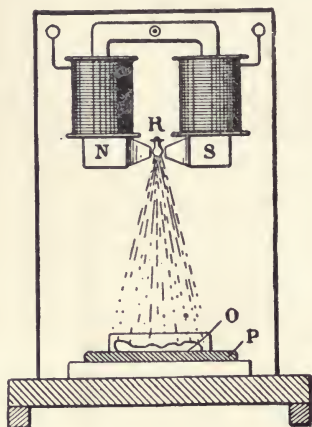


Fig. 33.

The figure illustrates the method for obtaining sharp radiographs at variable distances, with salts of radium. The ordinary radiographs made by radium are poorly defined on account of the diffusion of the  $\beta$ -rays. The  $\alpha$ -rays are absorbed by the container. The  $\beta$ -rays are got rid of through the influence of a powerful electro-magnet. The radium compound, R, therefore acts like a minute but powerful Crookes tube evolving Roentgen rays, which produce a skiagraph of the object, O, on the plate, P protected by black paper. The  $\gamma$ -rays from a small amount of radium may be caused to radiograph at variable distances, a meter or more, the time of exposure being much greater the farther apart are the plate and the source of energy.

1. C. R. 132, 545, (1901).

2. Proc. Roy. Soc. 72, 208, (1903).

cathode and  $\beta$ -rays; and that active products, giving off  $\alpha$ -rays and not  $\beta$ -rays, do not produce  $\gamma$ -rays. They appear to be very similar to the Röntgen rays that are produced in very "hard" tubes.

Radium preparations brought near other substances possess the power of inducing a secondary activity which diminishes at various rates. They do not appear to lose weight. Such conduct apparently questions the fundamental laws of physics and chemistry, namely the conservation of mass and energy. This will be dealt with in the fourth chapter.

Black<sup>1</sup> has shown that the electric resistance of selenium is diminished under the influence of radium rays. The action, though slower than in case of Röntgen rays, is of the same order of magnitude, as shown by Perrin. Van Aubel<sup>2</sup> has also shown that the electric conductivity of selenium is similarly affected in the neighborhood of hydrogen and oil of turpentine. Paillat<sup>3</sup> has called attention to the influence the radium rays have upon the electrical resistance of bismuth.

Gases subjected to the influence of radio-active substances, according to de Hemptinne,<sup>4</sup> became luminous under electric discharges or higher pressures than normal conditions. There is a similarity, although a difference, between the Röntgen and Becquerel rays, the red-violet color of the gas in the former becoming yellowish-green under the influence of radio-active bodies.

#### CHEMICAL ACTION OF RADIUM COMPOUNDS.

Radium preparations produce colors in glass, porcelain, rock salt, sylvite, etc. They have a destructive action on the skin (See Chapter V). Becquerel<sup>5</sup> converted yellow phosphorus into the red variety through the influence of the  $\beta$ -rays,

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1. C. R. 132, 15.
  2. C. R. 136, 929.
  3. C. R. 138, 139.
  4. C. R. 133, 934, (1901).
  5. C. R. 133, 709, (1901).

He, also, learned that mercuric chloride is reduced by oxalic acid when the mixed solutions are left in the dark with a radium tube. The germinating power of seeds is destroyed by an ante-planting exposure to the radiations.

Radium converts oxygen into ozone,<sup>1</sup> apparently through the influence of the  $\alpha$ - and  $\beta$ - and not the luminous rays.

Berthelot<sup>2</sup> in making a comparative study of specific chemical reactions caused by light, an electric current and radium, learned that, under the influence of the last named, iodic acid was decomposed, yielding free iodine. This did not occur when the radium was covered with black paper. Pure nitric acid was discolored at the end of a two days' exposure. These reactions are endo-thermic. Light caused the decomposition of carbon disulphide. Acetylene is readily polymerized by an electric current. Radium brought about neither of these exothermic reactions. Lead glass was turned black, manganese glass violet, hence the radiations were supposed to cause a reduction and oxidation simultaneously. However, later Ackroyd<sup>3</sup> showed that the color changes, as orange for sodium chloride, violet for potassium chloride, etc., produced by the  $\gamma$ -rays, corresponded to thermal effects in other bodies and are physical.

Mercurous sulphate, which darkens under the influence of ordinary light, especially the ultra-violet rays, is similarly affected by radium compounds.<sup>4</sup> The effect on the E. M. F. of a Clark cell is negligible, however.

Sudborough<sup>5</sup> has shown that certain labile stereoisomerides, as allo-cinnamic,  $\alpha$ - and  $\beta$ -bromo-allo-cinnamic acids are transformed into stable compounds more readily under the in-

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1. Curies, C. R. 129, 823, (1899).

2. Compt. Rend. 133, 18. Ann. Chem. Phys. (7), 25, 458, (1902).

3. "The Action of Radium Rays on the Halides of the Alkali Metals and Analogous Effects produced by Heat." Proc. Chem. S. 20, 108, (1904).

4. Skinner, "Action of Radium Rays on Mercurous Salts," Proc. Camb. Phil. Soc. 12, 260, (1904).

5. Proc. C. S. 20, 166, (1904).

fluence of light than by prolonged exposure to radium radiations.

Orloff<sup>1</sup> found that radio-active protuberances grew upon an aluminum plate exposed for three months above radium bromide in an ebonite capsule. He explained the phenomenon by the formation of a stable alloy with the accumulated material particles given off by the radium preparations.

Hardy and Willcock<sup>2</sup> found that a solution of iodoform in chloroform turned deep purple by resting the containing vessel on a sheet of mica covering radium bromide. The liberation of iodine from solutions of iodoform has been found to require oxygen and some form of radiant energy. The action was found to be due mainly to the  $\beta$ -rays, although the  $\gamma$ -rays produced the same effect. Röntgen rays produce a similar coloration.

Hardy exposed two solutions of globulin from ox-serum to the action of naked radium bromide. One solution was rendered electro-positive, by adding acetic acid; the other negative, by ammonia. The opalescence of the electro-positive preparation rapidly diminished, showing a more complete solution; while the electro-negative rapidly turned to a jelly and became opaque. This coagulation of globulin was found to be due to the  $\alpha$ -rays alone.

This unique substance, it has thus been seen, possesses properties that are most amazing. Although the trail has been followed close by many drawn to a contemplation of the wonder, it is safe to say that we perhaps are only on the threshold of a full knowledge of this marvel.

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1. Russ. Phys. Chem. Soc., April (1903).

2. Proc. Roy. Soc. 72, 200, (1903); Zeit. Phys. Chem. 47, 347.



## CHAPTER III.

OTHER RADIO-ACTIVE BODIES AND THE SOURCES  
OF RADIO-ACTIVITY.*Uranium.*

Becquerel<sup>1</sup> in his early studies of the invisible radiations emitted by salts of uranium attributed the darkening of the photographic plate to invisible phosphorescence. It did not seem to have any intimate association with visible phosphorescence or fluorescence. The sesqui-salts are fluorescent, while the uranous or green salts are not, yet the radiations from the latter were as intense as the former.

He<sup>2</sup> found that all uranium salts, as well as the metal, gave off invisible rays which penetrate gold, platinum and copper, black paper and affect a photographic plate.

Becquerel<sup>3</sup> also learned that the radiations from uranium and its compounds showed no appreciable variation after three years. The rays were absorbed in proportion to the thickness of any material they passed through. All the uranium rays deviate under the influence of an electro-magnet. They resemble Röntgen rays more than ordinary light. The radiating substances seem to be analogous to ordinary phosphorescent materials, but to retain relatively a very much greater reserve of energy.

Becquerel,<sup>4</sup> in an effort to concentrate the active body in uranium, treated its salts with barium chloride, and subsequently precipitated the latter by sulphuric acid. The precipi-

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1. Compt. Rend. 122, 689 and 762.
  2. Compt. Rend. 122, 1086.
  3. Compt. Rend. 128, 771 (1899).
  4. Compt. Rend. 131, 137 (1900).



tate carried with it a radio-active substance emitting rays deviated by a magnetic field. On repeating the operation eighteen times it was learned that the purified uranium possessed only one-sixth its original ability of ionizing air. Its rays passed more readily through glass than aluminum, whereas the converse was true for the original salt.

Crookes<sup>1</sup> learned that pure uranium nitrate fractionated with ether gave an inactive product (to the photographic plate), soluble in ether, while the activity became concentrated in the insoluble portion. He designated the active substance, provisionally, Ur-X. It differs from polonium, whose emanations do not pass through glass, aluminum or lead. It differs from radium in forming a readily soluble sulphate.

Becquerel<sup>2</sup> supposed uranium to contain a highly active body, probably actinium, as a strongly radiating body could be concentrated by adding a small portion of a soluble barium salt and precipitating with sulphuric acid. Yet the extreme products of a long series of fractionations of uranium nitrate by deBoisbaudran showed the same radio-activity, measured by the photographic and electrical discharge methods.

Becquerel<sup>3</sup> found that the temperature of liquid air, reduced the discharging power of uranium, determined by a very delicate electroscope, to about one-half of that noted at 25° C. Crystals of uranium nitrate plunged into liquid air or hydrogen became spontaneously luminous.

He<sup>4</sup> furthermore proved that the radio-activity of uranium was not constant, as Giesel had previously noted.

Soddy<sup>5</sup> repeated the work of Crookes. He directed attention to the fact, that by the photographic method, when the rays

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1. Proc. Roy. Soc. 66, 409 (1900).
  2. Compt. Rend, 130, 1583 (1900).
  3. Compt. Rend. 133, 4 (1901).
  4. Compt. Rend, 133, Dec. 9 (1901).
  5. Chem. News 86, 199 (1902).

are made to pass through cardboard or glass before reaching the sensitive film, only the  $\gamma$ -radiation will be measured, therefore the  $\alpha$ -radiation was left intact.

Rutherford<sup>1</sup> showed that the radiation from uranium was complex, the  $\beta$ -radiation being far more penetrating in character than the  $\alpha$ -radiation. The difficulty of making an accurate determination is due to the small conductivity produced by the  $\beta$ -radiation in the gas, as compared to that due to the  $\alpha$ -radiation.

#### *Thorium.*

In 1898 G. C. Schmidt<sup>2</sup> and Madame Curie<sup>3</sup> independently noted the radio-activity of thorium obtained from Bohemian pitchblende. Not long after the announcement of the Becquerel<sup>4</sup> rays, Crookes,<sup>5</sup> as noted above, showed that by fractioning uranium nitrate with ether, compounds could be obtained which did not affect the photographic plate. This indicated the separation of a new substance (Uranium X) and that radio-activity was not an inherent property of the element uranium, as maintained by Madame Curie.<sup>6</sup>

Soddy and Rutherford<sup>7</sup> demonstrated that only material carrying the  $\beta$ -rays was thus separated and that the inactive uranium (so called because it does not affect the photographic plate) still gives off  $\alpha$ -rays, which ionize gases and may be detected by the electrical method. Crookes, in the same paper, reported a few preliminary experiments on thorium compounds and suggested "the possibility of separating thorium from its radio-active substance."

Hofmann and Zerban<sup>8</sup> found that the activity of thorium could be fractioned away. The activity is increased in that

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1. Phil. Mag. Jan., 1897.
  2. Wied. Ann. 65, 141.
  3. Madame Curie's Thesis, Faculté des Sciences de Paris (1903).
  4. Compt. Rend. 122, 420, 501, 559, 689, 762, 1086 (1896).
  5. Proc. Roy. Soc. 66, 406 (1900).
  6. Compt. Rend. 127, 175.
  7. Proc. Chem. Soc. 18, 121.
  8. Ber. d. chem. Ges. 35, 531 (1902).

portion most readily precipitated by potassium sulphate, chromate, hydrogen dioxide, and sodium thio-sulphate. With ammonium carbonate the more active portion passes into solution. They also examined a number of minerals from which thorium is obtained and proved the presence therein of uranium. The thorium oxides from all of these were radio-active. Norwegian gadolinite, orthite and yttrotitanite free from uranium gave a thorium oxide which neither affected the electroscope nor the photographic plate.<sup>1</sup>

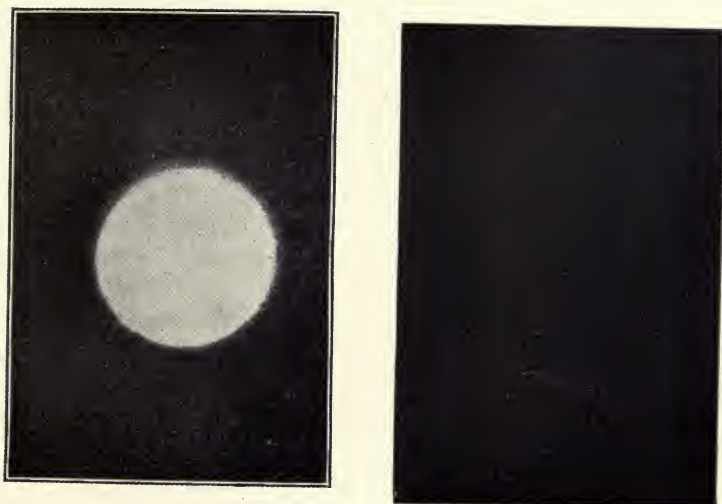


Fig. 34.

This illustration shows on the left the action of a strong radio-active thorium preparation acting through 1 m.m. of glass upon a plate; on the right the same preparation through 1 m.m. of glass was placed, a half year later, black paper only protecting the powder from the plate. The loss of the induced(?) activity, or at least that portion affecting a silver-bromide gelatine preparation is very noticeable. The exposure was for twenty-four hours in both cases (After Zerban.)

1. Ber. d. Chem. Ges 35, 533 and 145 (1902).

The work of Hofmann and Zerban touching the primary activity of thorium being questioned by Barker,<sup>1</sup> was upheld by the junior author who detected the presence of uranium in monazite.

The elegant researches of Rutherford and Soddy<sup>2</sup> proved that there can be no doubt of the existence of a novel highly radio-active substance with thorium (thorium X), as it is usually extracted from minerals without consideration of their chemical composition. Hofmann and Zerban strenuously direct attention to this last fact. Such prepared so-called pure salts of thorium contain a radio-active constituent, which may be concentrated chemically by precipitation with ammonia (the filtrate carries thorium X)<sup>3</sup> and washing the oxide with acid or even water. The residues obtained by evaporation of the ammoniacal solution in the first case are a thousand times as active as the original and "are free from thorium, or, at most, contain only the merest traces, and when redissolved in nitric acid do not appear to give any characteristic reaction." The residue from the water washings became 1,800 times as active, and after conversion into sulphate, Rutherford and Soddy state, "*No other substance than thorium could be detected by chemical analysis*, although, of course, the quantity was too small for a minute examination"<sup>4</sup> (See Emanium).

Giesel<sup>5</sup> said the radio-activity of thorium could not be due to actinium.

When we consider that barium chloride containing radium may be precipitated by sulphuric acid or silver nitrate and the filtrate or precipitate obtained thereby, supposedly containing none of that remarkable body, is still radio-active,<sup>6</sup> we can easily

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1. "The Radioactivity of Thorium Minerals," Am. J. Sci. 16, 164 (1903).

2. Proc. Chem. Soc. (London), 18, 2 (1902).

3. Rutherford and Soddy; Phil. Mag. (1902), p. 370.

4. Italics theirs.

5. Berichte 34, 3776.

6. See the works of the Curies, Giesel, Elster and Geitel, Marckwald and others.

understand how in a mineral or salt a radio-active body, perhaps resembling one of the constituents, clings to various components throughout many chemical manipulations. It having been suggested that uranium might owe its radio-activity to the presence of small amounts of polonium or radium, Mme. Curie<sup>1</sup> states that such could not be true, and adds in another paper,<sup>2</sup> "the property of emitting rays, \* \* \* \* which act on photographic plates, is a specific property of uranium and thorium." "The physical condition of the metal seems to be of an altogether secondary importance." "Uranium and thorium alone are practically active."

The power possessed by thorium, as usually prepared, of inducing activity, reported by Rutherford and his co-workers,<sup>3</sup> is most interesting. The brilliant French woman states concerning uranium:<sup>4</sup> "I have never found any marked difference between the relative activities of the same compounds." By analogy one may consistently assume the same for thorium. The author has obtained similar compounds of thorium fractions which do differ in their radio-activity, in some cases one being three times as great.

Metzger<sup>5</sup> has published an interesting and novel method for separating thorium from cerium, lanthanum, and didymium, depending upon its precipitation from a neutral solution by a forty per cent. alcoholic solution of fumaric acid. The writer and Lemly<sup>6</sup> have verified these observations as far as ordinary analytical methods are concerned. Applying it to the accepted chemically pure thorium, however, we obtained a filtrate containing less than 0.5 per cent. of the original, which, on evaporation and ignition, gave a grayish oxide possessing such marked

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1. *Revue générale des Sciences*, January, 1899; M. and Mme. Curie: *Compt. Rend.*, 127, 175.

2. M. and Mme. Curie and M. Bemont: *Compt. Rend.* 127, 1215.

3. *Loc. cit.*

4. *Ibid.*

5. *Journ. Amer. Chem. Soc.* 24, 901.

6. Unpublished work.





radio-activity by the electrical method that Dr. Pegram stated, that it acted as if "salted with radium." This decayed with fair rapidity, from 42 to 12.4 in eight days, to 3.3 in nine days more (uranium being taken as the standard unit). After thirty-two days more it gave 3 and was practically constant. The corresponding values obtained for the thorium precipitate, which constituted virtually the whole, were 0.63, 0.92 and 1. Truly but one interpretation of these results may logically be had, namely, the existence of a radio-active body with thorium, which is different from it. Crookes has sounded a timely warning against depending upon the photographic method for determining the radio-activity, so we have been guided mainly by the electrical method. Although by no means comparable to the other procedure, yet most interesting observations may be had by the photographic method. It has been used to secure very rough quantitative results. It aids one much in learning of the  $\beta$ -activity. The  $\alpha$ -rays are the most important factor in the ionization of gases, upon which depends the electrical method. Persistent differences, in radio-activity of the preparations had by different chemical methods, have been noted and the same method of preparation has given persistent differences in radio-activity measured by the same and different methods.

Radio-active thorium obtained from monazite has been resolved by the writer into at least two and most likely three different constituents.<sup>1</sup> All these are radio-active, but of different strength. Recently the writer and Zerban have obtained a thorium preparation from an inactive South American mineral, which is free from any activity whatever.

Ramsay reported the extraction of a very active body like thorium from a new Ceylon mineral. This cubical mineral is very radio-active and gives out large amounts of helium, which Tyrer has collected in twenty-five liter lots. The constituent,

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1. Thorium; Carolinium, Berzelium. Journ. Amer. Chem. Soc. 26, 922 (1904).

resembles thorium and shows an atomic weight of 240 in the impure form and may contain the carolinium of Baskerville.

*Polonium.*

When pitchblende was dissolved in acid, and sulphuretted hydrogen added, the sulphides obtained were very active. On purification, a substance similar to bismuth was obtained and Madame Curie<sup>1</sup> named this first radio-active element *Polonium*, after her native country.

Polonium may be partially separated by any one of the following three methods: First, the active sulphide, being more volatile than bismuth, may be sublimed in a tube between 250° and 300° C. and the active body is obtained as a black substance. Second, the active sulphide is less soluble than the inactive one, hence partial separation may be obtained by precipitation with sulphuretted hydrogen in a hydrochloric acid solution. Third, polonium nitrate hydrolyses more easily than bismuth nitrate, therefore the active body is precipitated by adding water to the nitric acid solution.

By using the last mentioned method, which is very slow and tedious, Madame Curie<sup>2</sup> obtained a small quantity of material extremely active compared with uranium. Only the bismuth lines were observed in the spectrum, as reported by Demarçay, Runge and Exner. Crookes<sup>3</sup> observed one new line in the ultra-violet and Berndt<sup>4</sup> saw a large number of new lines in the same region when he used a polonium of 300 activity.

Polonium apparently gives radiations that are very easily absorbable. Giesel<sup>5</sup> called attention to the fact that the penetrating power of the polonium radiation is much less than that of radium rays, consequently the shadow produced by an object

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1. C. R. 127, 175 (1898).

2. See her thesis.

3. Proc. Roy. Soc., May (1900).

4. Phys. Zeit. 2, 180 (1900).

5. Ann. Phys. Chem. (1899), 11, 69.

is much sharper and deeper with the former than with the latter (Fig. 35).

Becquerel examined samples of polonium nitrate nearly as active as the radium salts then had. The radiations appeared to be unaffected by the magnetic field, thus differing from those of radium.

The activity is not constant, but diminishes regularly according to the time. In eleven months Madame Curie found that polonium lost one-half of its original activity. This fact has caused many to view polonium not as a new element, but merely as active bismuth, for it is well known that inactive elements in the presence of active ones acquire activity. There is another view of the matter, however; namely, the supposed presence of a very small quantity of some intensely active matter. Polonium is therefore not as yet accepted as an element.

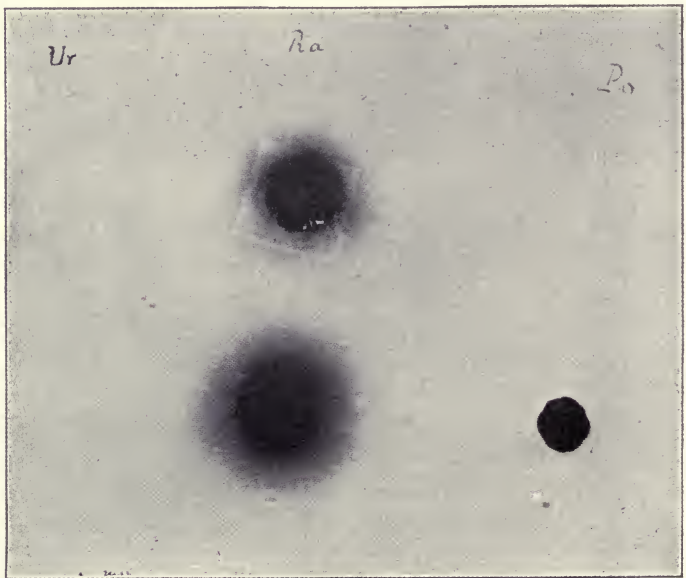


Fig. 35.

According to Becquerel, polonium rays do not pass through a thin film of black paper, forming a small cylinder enclosed with aluminum or mica, upon the bottom of which rests the powder; the radium rays readily transverse the envelope. The figure illustrates strikingly the difference in the rays.

Marckwald,<sup>1</sup> entering the discussion as to the elementary character of polonium, reported the finding of a substance in pitchblende similar to polonium, but whose activity did not decay with time. A rod of bismuth dipped into the active solution obtained from the uranium residues quickly acquired a black, intensely active deposit. (See radio-tellurium.)

Giesel<sup>2</sup> confirmed Marckwald's statement that bismuth immersed in a solution of Curie's polonium, acquired the property of emitting  $\alpha$ -rays. Bismuth, platinum, and palladium may be rendered highly active by immersion in a solution of radium salts. After the metal is carefully washed with hydrochloric acid and water, to remove traces of radium, it still emits the  $\alpha$ -rays strongly. Bismuth becomes much more active than the other two elements, consequently Giesel insisted that polonium is nothing more than bismuth rendered active by contact with radium salts. This German scientist also learned that after bismuth remained in a one per cent. radium salt solution for several days and was then removed and washed with hydrochloric acid and water, it showed intense  $\alpha$ -radiation, but no  $\beta$ -radiation. The small quantities of bismuth and platinum metals that were dissolved in this experiment were precipitated with hydrogen sulphide. These sulphides were found to emit the  $\beta$ -rays. This may be ascribed to the adhering radium salts. One of the methods for distinguishing the different rays depends upon the observations of Crookes, Elster and Geitel; namely, the scintillations produced by the  $\alpha$ -rays, whereas the  $\beta$ -radiations produce only uniform illumination of the screens.

The Curies,<sup>3</sup> from various observations, concluded that the radio-activity of uranium, thorium, radium, and probably actinium, is the same and does not vary with time when the radio-active substance is brought into the same chemical and physical state. If by any particular treatment the substance loses any of its activity, it regains it in the course of time. Polonium acts differently.

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1. Berichte 35, 2285 (1902) and elsewhere.

2. Berichte 36, 23, 68 (1903).

3. Compt. Rend. 134, 85 (1902).

*Actinium.*

Debiere,<sup>1</sup> who directed the factory work of the Curies, obtained from pitchblende an active substance which was precipitated in the iron group and which appeared to be very closely allied to titanium and thorium, especially the latter. He named it *Actinium*. This substance has never been obtained in sufficient purity to give a characteristic spectrum. It should be mentioned, however, that the spectrum of thorium itself is extremely complicated. Four methods were used by Debiere for partial separation, as follows:

First. The active matter accumulated in the precipitate produced by sodium thio-sulphate in hot solutions, made slightly acid with hydrochloric acid.

Second. Titanium was separated by the action of hydrofluoric acid upon the suspended hydrates in water. The actinium accumulated in the undissolved portion.

Third. When a neutral nitrate solution was precipitated by hydrogen peroxide, the active body accumulated in the precipitate.

Fourth. If the sulphates were treated with a soluble barium salt, the barium sulphate which was formed carried down the actinium matter. This was separated from the barium after conversion into the chloride, by cooking with sodium carbonate, filtering, dissolving the precipitate in hydrochloric acid and adding ammonia.

The original material was free from uranium. After separating the other known radio-active bodies, radium and polonium, a preparation 5,000 times as active as uranium was obtained.

Actinium renders gases capable of discharging electrified bodies, excites fluorescence in a barium-platino cyanide screen and affects photographic plates (Fig. 36). It is said to differ from radium in not being spontaneously luminous.

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1. C. R. 129, 593 (1899); 130, 906 (1900).



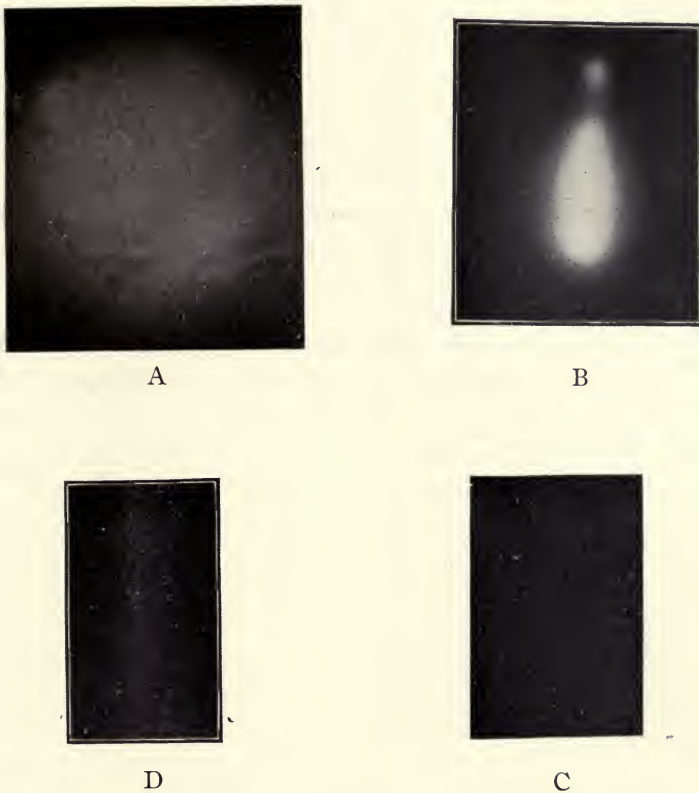


Fig. 36.

The figure illustrates the action of radium bromide 300,000, "uranies" strong, (A and B) and actinium oxide, 10,000 uranies, (C and D) on photographic plates through the thin glass of the containers. A and C were exposed to the plates, covered with black paper, for two hours. Practically no action on the sensitive film was observed for the actinium. B and D were allowed to remain fourteen hours. A marked difference is observed in the character of the radio-activity of the two substances. That of radium is more complex. The term "uranies" was coined by Roberts to mean the standard in terms of the activity of metallic uranium (unity).

Actinium gives out easily absorbable and penetrating, deviable rays, like cathode streams, and a radio-active emanation, which loses its activity in a few seconds. While it resembles the thorium emanation, it differs from it in the rate of decay.

The thorium emanation loses one-half of its activity in one minute.<sup>1</sup>

The deviation<sup>2</sup> was found to correspond to positively charged bodies moving with a high velocity. The induced radio-activity was shown by Debierne in the following manner: Two plates were placed at an angle over a tube containing an actinium salt. The ions are contained almost exclusively in the tube above the salt, but the plates become radio-active. This can only be accounted for by a secondary radiation proceeding from each ion, as the radiation is deviated in a magnetic or electric field as has been established.

No atomic weight has been obtained for actinium. Hofmann and Zerban<sup>3</sup> obtained the equivalent 63.32 or atomic mass value of 253.28 (tetrad.) for a preparation of 2,000 activity.

#### *Radio-Active Lead.*

Elster and Geitel<sup>4</sup> obtained a very active lead sulphate from pitchblende. They were able to extract from this an inactive lead sulphate. Therefore, they attributed its activity to the presence of more or less radium.

Giesel<sup>5</sup> obtained a small sample of radio-active lead sulphate from uranium residues which, when wrapped in black paper, did not produce any effect upon the photographic plate. When surrounded by transparent paper, however, the plate was affected. He, therefore, attributed the action to that of light rays given off by the phosphorescent substance. He calls attention to the fact that lead might contain a very minute amount of radium, much less than one is able to detect by chemical means, and still be radio-active.

Hofmann and Strauss<sup>6</sup> obtained a lead sulphate from pitchblende, uranium, mica, bröggerite, cleveite, and samarskite,

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1. C. R. 136, 446 (1903).
  2. Compt. Rend. 136, 671 (1903).
  3. Berichte 36, 3093 (1903).
  4. Wied. Anal. 69, 83 (1899).
  5. Berichte 34, 3772 (1901).
  6. Berichte 33, 3126 (1900).

which were radio-active. It contained no trace of bismuth, barium, (which precluded the presence of polonium and radium), titanium, thorium, or uranium. The sulphate was insoluble in dilute sulphuric acid, but easily soluble in ammoniacal tartrate. The chloride showed diminished radio-activity when crystallized from hot water, whereas the mother liquor gave crystals with increased activity.

These same workers<sup>1</sup> purified their active lead sulphate by converting it into the carbonate and then into the chloride. They were able to fraction the sulphate into more active preparations, which gave a blue phosphorescence when exposed to the cathode rays. The spark spectrum gave a characteristic line in the violet.

Potassium iodide converts the sulphate into a mass of yellow crystals, which dissolve in warm dilute sulphuric acid and separate again on heating. From a determination of the percentage of the sulphur tetroxide, it was learned that the metal present was both bivalent and quadrivalent. The radio-activity of the sulphate diminished on keeping, but was entirely restored on exposure to the cathode rays. The atomic weight of 260 has been assigned to radio-active lead. It resembles ordinary lead in most of its characteristics, except that the sulphate is strongly phosphorescent.

They<sup>2</sup> also isolated from bröggerite two elements (?) of the atomic weights 100.92 and 171.96. The former gives a yellow sulphate and has little influence on the radio-activity of the lead. The strong radio-activity of the other is lost when converted into the sulphide. This is obtained again when the sulphide is reconverted into the sulphate.

The chromate<sup>3</sup> of the radio-active lead is not decomposed on repeated warming with dilute sulphuric acid, which distinguishes it from ordinary lead chromate. The sulphate acts

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1. Berichte 34, 8 (1901).

2. Berichte 34, 907 (1901).

3. Berichte 34, 3033 (1901).

upon a photographic plate through aluminum and glass. All of the radio-activity is effective in discharging the electroscope.

Hofmann with Wolff<sup>1</sup> found that the radio-active lead could be concentrated by dissolving the lead sulphide in aqueous sodium thio-sulphate. On keeping for several days an active black sulphide separated out. Unlike polonium, this radio-active lead acted on a photographic plate with great rapidity through a sheet of gutta percha.

Giesel<sup>2</sup> found that the radio-activity of the radio-active lead which he obtained diminished with time, whereas Hofmann's preparations preserved their activity.

Winkler has questioned the method of the determination of the atomic weight, but Hofmann has apparently substantiated his claims. (See Chapter V.)

#### *Radio-Tellurium.*

Marckwald<sup>3</sup> obtained 1.5 grams of radio-active tellurium from six kilograms of bismuth oxychloride which was extracted from 2,000 kilograms of pitchblende. This contained only about one per cent. of the radio-active constituent. The whole was converted into the chloride and precipitated by hydrazine. The filtrate was concentrated and heated on a water bath with a drop of stannous chloride. In this way four milligrams of a dark colored precipitate were obtained. This radio-active substance dissolves in cold nitric acid and may be converted into a soluble chloride. Upon the immersion of a copper, tin, or antimony plate in the solution, the active substance is deposited in a fine state of subdivision. 0.01 milligram separated on the copper plate. Four square centimetres of this plate illuminated a zinc-blende screen so that it was visible to several hundred people.

The polonium of Madame Curie behaves quite differently from radio-tellurium, in that its nitric acid solution gives with

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1. Berichte 35, 1453 (1902).

2. Berichte 34, 3775 (1901).

3. Berichte 36, 2662 (1903).

water a yellow to brown precipitate, soluble in acids. The polonium of Giesel is also quite different.

Bismuth dipped into a solution containing radium becomes radio-active, but its activity is not comparable with that of radio-tellurium. The solution is not at all exhausted.

Tellurium precipitated by stannous chloride from a solution of tellurous acid, containing radium chloride, although somewhat active, gives when converted into the chloride, a liquid which fails to render active a copper strip immersed in it. Therefore, the induced activity is doubtless different from the activity of radio-tellurium.

Marckwald,<sup>1</sup> who entered a discussion on the complicated question of the nature of polonium, secured from pitchblende a substance resembling tellurium, which is active, but whose activity does not decay with time. The method of separation depended upon the insertion of a rod of bismuth into the bismuth chloride solution, obtained from the uranium residues. The black deposit which coats the bismuth is very active. Having obtained .6 gram of the substance, he proved that its activity did not decay within nine months.

The chloride was electrolysed for three days; a bismuth cathode and carbon anode were used. The solution became inactive. The deposited metal is much more radio-active than the original substance. The deposit, which contains a little chloride, was sublimed from the bismuth terminal. The metallic bead obtained was dissolved in nitric acid. The solution gave the reactions of bismuth.

The rays given out did not penetrate paper or other obstacles, hence they were similar to those given out by polonium. They affected a photographic plate, and caused diamonds, zinc oxide and other substances to fluoresce brightly. The body differed from polonium in the fact that its activity did not decay.

This method of producing radio-active metallic coatings has been patented by H. H. Lake, of the firm of Stahmer & Co., Hamburg.<sup>2</sup>

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1. Ber. Chem. Ges. 35, 2285 (1902), Chem. Zeit. 26, 895.

2. J. S. Chem. Ind. 22, 1136 (1903).



According to the periodic table of Mendelejeff, tellurium should have an atomic weight less than iodine, whereas it has actually been found to be greater. Pellini<sup>1</sup> suggested that this might be accounted for by the presence of a small quantity of some element, which has a higher atomic weight (about 212), similar to tellurium and analogous to the radio-active constituents of pitchblende.

#### *Emanium.*

Giesel<sup>2</sup> has obtained from pitchblende a substance which he has termed *Emanium*. It appears to be allied to lanthanum, belonging to the cerium group of rare earths. The salts when first prepared are not so active, but reach a maximum in about a month, remaining so indefinitely; thus being similar to radium.<sup>3</sup> The original  $\beta$ -radiation becomes smaller the longer the substance is kept in solution. Runge and Precht obtained a number of new lines in the spectrum, which gave essentially the lines of lanthanum and a little cerium. The lines of thorium, barium, and radium were not present. The anhydrous chloride and bromide phosphoresce spontaneously. Glass vessels in which the substance is kept for a month become violet colored; paper is turned brown and destroyed.

"If solutions of thorium, lanthanum, cerium, etc., to which radium has been added, are precipitated with ammonia and washed, the precipitates are adulterated with traces of radium and show, besides  $\beta$ -radiations, remarkably strong  $\alpha$ -radiations, but yet an emanation similar to the emanating substance."<sup>4</sup>

The discoverer attributes the induced radio-activity of many substances rather to the emanating substance than to the presence of minute traces of radium.

A splendid experiment, visible a great distance, may be performed by blowing a current of air through tubes containing the material against a large screen of Sidots blende. The

1. Gazetta 33, 11, 55 (1903).

2. Berichte 35, 3608 (1902); 36, 342 (1903).

3. Ann. d. Phys. u. Chem. 69, 92 (1899).

4. Giesel, Berichte 36, 342 (1904).

scintillations are most pronounced and are visible to the naked eye, representing a large spinthariscopes.

In studying the radio-activity of thorium, at the request of the writer, Dr. H. S. Miner, of the Welsbach Lighting Company, saved certain ammoniacal washings obtained in the process for the extraction and purification of thorium oxide from monazite sand for the manufacture of gas mantles. The ignited residue, obtained from evaporating over 100 liters of this liquor, produced a marked effect upon the photographic plate and showed nearly three times the radio-activity of thorium by the electrical method, using the apparatus of Dolezalek. The radio-activity remained constant through a number of months. The body gave none of the chemical reactions and did not show a single line of thorium in the arc spectrum.

The writer, working with Lichtenthaeler, has obtained highly radio-active bodies, tested by the photographic method, from thorium, cerium, didyium oxides, and the residual phosphates, extracted by ourselves from North Carolina monazite sands. Further, we obtained an extremely active body by precipitating the sulphate solution with hydrogen sulphide, which perhaps would, but not necessarily, indicate the presence of polonium. Apparent verification is thus had of Giesel's work on emanium.

E. Goldstein<sup>1</sup> examined Giesel's emanium as obtained from pitchblende and in its chemical behavior it seemed to be related to cerium. On account of the small penetrative power of the emanation, he assumed that the air exerted a strong absorption of the latter so that its effects would be augmented in exhausted tubes. This was verified by experiment. His experiments indicated that the observed luminescence is due rather to a gas, than a special form of energy issuing from the substance. When cooling by means of liquid air exhausted tubes, where the active matter had been introduced, Goldstein observed a very strong luminescence on the wall, which, instead of rising in the closed

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1. German Physical Society.

portion of the tube, seemed to be confined to the zone immediately above the level of the liquid air. That is, the phenomenon is characteristic of a definite temperature above the temperature of liquid air. The emanation is given off at the temperature of liquid air. He does not think that the emanation energy in question is identical with that of radium, the distinguishing features being first, the absence of the coloration of the tubes, and, second, the excessively low penetration.

*Artificially Active Barium.*

Debiegne<sup>1</sup> observed that when a highly radio-active salt of actinium is added to a solution of barium chloride, the latter becomes radio-active. If the barium be precipitated as sulphate and reconverted into the chloride, the actinium being separated by means of ammonia, the barium retains some activity. This may be increased, depending upon the time of contact, till it is several hundred times as active as uranium. It is persistent throughout various chemical changes, it ionizes gases, excites barium-platino-cyanide, and acts upon a photographic plate. Part of the radiation is deflected in a magnetic field and the anhydrous chloride is also luminous. This barium chloride, rendered radio-active, may be fractioned similarly to the radium salts. The salts, however, do not give any of the lines of the radium spectrum and their radio-activity gradually diminishes in intensity.

#### THE NUMBER OF RADIO-ACTIVE ELEMENTS.

Soddy<sup>2</sup> maintains that radio-tellurium is identical with polonium and that there is no justification for the assumption of a new element. Soddy acknowledges the existence of five radio-active elements; namely, uranium, thorium, polonium, radium, and actinium. They may be distinguished in three ways: They all give off  $\alpha$ -rays; all, with the exception of polonium, give  $\beta$ -rays; uranium, thorium, and radium give

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1. Compt. Rend. 131, 333 (1900).

2. Nature, March 17, 1904, pp. 461.

$\gamma$ -rays. Polonium does not, and it is questionable about actinium. Neither uranium nor polonium gives off a radio-active emanation, while thorium, radium and actinium do. Those radio-active substances which give off emanations impart activity to surrounding objects. That is, substances placed in the neighborhood of thorium, radium and actinium acquire an activity which is not permanent. The three substances which give off emanations have their respective emanations distinguished from one another by the time their activity lasts. The emanations of radium continue through several weeks, those of thorium only a few minutes, and actinium only a few seconds.

#### OTHER SOURCES OF RADIO-ACTIVITY.

C. T. R. Wilson<sup>1</sup> reported the radio-activity of rain and snow. Rutherford and Allan<sup>2</sup> studied excited radio-activity and its effect on the ionization of the atmosphere. The latter regarded the radio-activity of rain and snow as derived from the radio-activity of the atmosphere. J. J. Thomson<sup>3</sup> reported a radio-active gas in the Cambridge tap water, as did Bumstead and Wheeler<sup>4</sup> for the surface water around New Haven, Conn. Adams,<sup>5</sup> considering the former, suggested the presence of a small amount of radium in solution. Knett<sup>6</sup> observed that the thermal springs of Karlsbad deposited small yellow tabular crystals of barium sulphate which were very radio-active.

McLennan and Burton<sup>7</sup> reported the electric conductivity of the atmosphere. The former<sup>8</sup> has observed the radio-activity

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1. Proc. Camb. Phil. Soc. 12, 17 (1902); 13, 85 (1902).

2. Phil. Mag. 6, 704 (1902).

3. Proc. Camb. Phil. S. 123, 172 (1903).

4. Am. J. Science (1904).

5. Phil. Mag. (6), 6, 563.

6. Sitz. Wien. No. 11 (1904). Nature 70, 160.

7. Phil. Mag. 6, 5, 699.

8. Nature 70, 151 (1904).

of natural gas, as well as experimented<sup>1</sup> on the induced radio-activity excited in the air at the foot of waterfalls. McLennan and Burton<sup>2</sup> also learned that metals generally possess more or less radio-activity.

H. Lester Cook<sup>3</sup> reported the penetrating radiation from the earth's surface. Borgmann<sup>4</sup> found the Russian muds radio-active and that the air could be electrified by metals. Geitel found a wire electrically charged and suspended in the air, as well as the ends of pine needles, radio-active.

Elster and Geitel<sup>5</sup> secured a radio-active emanation from the air, from the soil, from cellars, mountain tops, mines, etc. They<sup>6</sup> also observed the radio-activity of sediments obtained from evaporated spring water.

Müller<sup>7</sup> verified these observations and suggested the presence of another radio-active element accompanying radium.

Strutt<sup>8</sup> studied the properties of a strong radio-active gas, which he obtained from metallic mercury and learned that the emanation behaved similar to that of radium, reaching one-half value in a little over three days. Strutt<sup>9</sup> also determined the activity of a number of minerals, mineral waters, and ordinary materials. Lester Cooke<sup>10</sup> proved the universal occurrence of a penetrating radiation similar to radium. It may have its origin in the radio-active matter distributed throughout the earth and atmosphere. Himstedt<sup>11</sup> reported the radio-active emanation of

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1. Phil. Mag. 6, 5, 419.

2. Phil. Mag. 6, 6, 343 (1903).

3. Phil. Mag. 6, 6, 403 (1903).

4. Nature, 70, 80 (1904).

5. Chem. News 88, 29 (1903).

6. Phys. Zeit. 5, 321.

7. Phys. Zeit. 5, 367.

8. Phil. Mag. 6, 6, 113.

9. Proc. Roy. Soc. 73, 191; Phil. Mag. (6), 5, 680.

10. Phil. Mag. 6, 410 (1903); Proc. Roy. Soc. 68, 151; 69, 277; Nature (1903), 369; 391, 414, 439.

11. Ann. d. Phys. 13, 573.



water and oil springs, and von Traubenberg<sup>1</sup> considered the absorption of the emanations of radium by the tap water of Freiburg and other liquids.

Different meteorological conditions<sup>2</sup> appear to determine different degrees of radio-activity of the air. Much activity is excited in fog. In cold, frosty weather the activity of the air is very high. We have learned that tobacco smoke in the room where one is making measurements by means of an electro-scope increases the conductivity of the air. These things cause variations in the leak of the instruments.

Concerning the general radio-activity of metals, Voller<sup>3</sup> has called attention to a flaw in the experiments by McLennan and Burton,<sup>4</sup> who claimed to be able to prove all metals radio-active, as the potentials are very small and subject to many errors. Hallwachs has pointed out the necessity of taking into account all the E. M. F.'s of the electro-metric system. But Voller, on the other hand, says it cannot be denied that the spontaneous projection of positive ions by all metals, if conclusively established, would mean a very important advance in our knowledge of the electrical phenomena.

Himstedt<sup>5</sup> arrived at the conclusion that radio-active bodies give off gaseous emanations and are widely diffused throughout the earth. These emanations, being absorbed by water or petroleum, are afterwards conveyed along by the latter to the surface of the earth and are diffused into the air. On account of the analogy between these emanations and those of radium, he puts forward the belief that they are identical. The ores of uranium, from which the radium emanations are derived, are therefore either widely diffused or there are other substances possessing, perhaps to a lower degree, the property of giving off emanations. The absorption co-efficient of water

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1. Phys. Zeit. 5, 130 (1904).

2. London Lancet, Aug. 8th (1903).

3. Phys. Zeit. Oct. 1 (1903).

4. Electrician, Sept. 11, 1903, p. 839.

5. Phys. Zeit. Apr. 15 (1904).

and petroleum, with respect to the emanations, is found to decrease with the increase of temperature.

Hot fountains have been found which show especially high activity. The hypothesis is, therefore, put forward that the amount of radio-active material increases for augmenting depths. The radio-active components of the earth, consequently, should have to be allowed for in accounting for the temperature of the earth.

Schuster,<sup>1</sup> referring to the matter of the cosmical radio-activity, calls attention to the fact that any physical property discovered in one element has always been found to be shared by all. The possibility that radio-activity may be a common property of all matter is immediately suggested. Radio-active bodies, therefore, may be distinguished from other bodies by the enormously exaggerated form in which they possess the property.

We know that the earth must be charged with negative electricity, which must be constantly renewed as there is constant leakage. Elster and Geitel recently determined that a body loses about 1-1/3% of its charge per minute. If the air near the ground has that conductivity, the earth should lose about one-half of its charge in an hour. There can be little doubt, therefore, but that we are living in an electric field through which negatively charged particles are constantly driven and which possesses an electric conductivity similar to that found in the neighborhood of radio-active bodies. The radio-activity of the ground air or water may thus be the consequence of the emanations of a radio-active earth.

Schuster also pointed out the possibility of a corresponding radio-activity of the matter in celestial bodies.

Rutherford, in a lecture before the Royal Institution in London, stated that the amount of radium present and uniformly distributed throughout the earth would be sufficient to account for all the heat lost from that body. On the assumption that this is true, the period of time for the cooling of the earth till it becomes uninhabitable, as calculated by Lord Kelvin, may be extended a few million years.

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1. Chem. News 88, 166 (1903).

## CHAPTER IV.

RADIO-ACTIVE EMANATIONS AND SECONDARY  
RADIO-ACTIVITY.

Variations in the radiations of thorium have been observed, by the electrical method, when the substances were examined in open vessels. Owens<sup>1</sup> found that this was caused by the air currents. When active matter was introduced into a closed glass vessel the activity increased with time, finally reaching a constant, which could be reduced by passing a stream of air through the vessel. It was noted, also, that the radiations passed through several thicknesses of paper, which absorbed the  $\alpha$ -rays. Rutherford<sup>2</sup> discovered the emission of radio-active particles from thorium compounds and named them "emanations."

The ionizing substance acts upon a photographic plate and diffuses through porous substances similar to a gas. (Fig. 36.) It may be swept along by a current of air, passed through cotton-wool, and bubbled through a solution of caustic potash without any loss of activity. Thus it differs from the ions produced in gases by the action of radio-active substances. The emanation cannot be dust of radio-active substances, which would be screened out by the cotton-wool filters. Hydrogen peroxide possesses the power of diffusing rapidly through porous substances and acting upon a photographic plate, but it is not radio-active. It is the radiation from the emanation, and not the emanation itself, that produces the two characteristic ionizing and photographic effects. (Fig. 37.)

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1. Phil. Mag., Oct. (1899).

2. Phil. Mag., Jan. (1900).

To Rutherford and his co-workers are we indebted for most of our knowledge of these and other emanations. Dorn<sup>1</sup> made similar discoveries later with radium compounds. The radium emanation loses its radio-activity at a different rate, although it possesses many similar properties. Both behave like a temporarily radio-active gas mixed in minute quantities in the air in which they are conveyed.

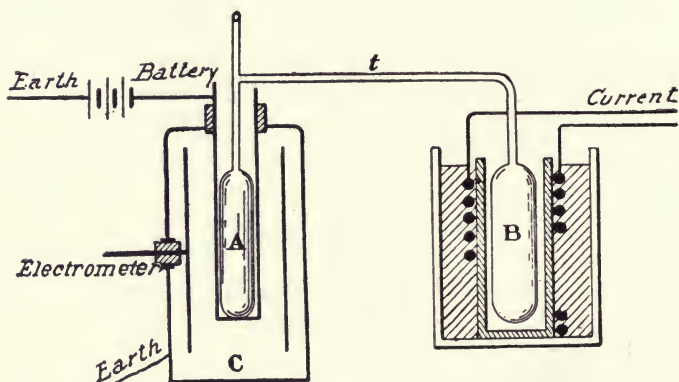


Fig. 36.

The apparatus may be used to demonstrate that the emanation of radium is a gas and follows Gay-Lussac's law. The bulbs, A and B, connected by a glass tube are evacuated, filled with the emanation and placed as shown in the diagram. A rests within a cylindrical condenser, such as shown in Fig. 36, while reservoir B rests within a constant temperature bath which may be heated by electricity. The radiation of A possesses a definite value for one temperature, increasing with the elevation of the temperature of B. The quantity of the emanation which has been driven out corresponds to that which it should be according to Gay-Lussac's law.

The emanations are given off more generously by the radium compounds by heating, or on solution in water. Curie

1. Abh. d. Naturforsch. Ges. für Halle (1900).

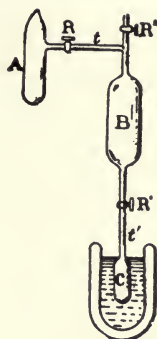


Fig. 37.

The figure shows apparatus, which may be used to illustrate the diffusion and condensation of the emanation, in short demonstrate its gaseous nature. Bulb A contains a radio-active solid or its solution. B is coated on the inside with Sidot's blende, as is also the small bulb C. Inserted in the connecting tubes, t and t' are the glass cocks R and R'. R'' serves to disconnect the apparatus from a vacuum pump. The emanation collects in A, R being closed, if the radio-active solution be allowed to stand; if a solid be used, the experiment may be hastened by gently heating the bulb. B and C are evacuated, R'' closed. On opening R, the emanation which has caused a slight glow in A, flows into B, which becomes quite luminous. On closing R, placing C in a Dewar bulb containing liquid air and opening R', the emanations are drawn into C; after a time close R' and remove the apparatus. B will have ceased glowing and C is exceedingly luminous in a dark room.

and Debierne<sup>1</sup> found that if radium preparations were placed in a vacuum tube, the vacuum was continually weakened. Giesel<sup>2</sup> observed that gases were evolved from solutions of radium bromide, which Runge and Bödlander found, by spectrum examination, to be mainly hydrogen with about one-tenth oxygen. Ramsay and Soddy<sup>3</sup> found that 50 m.g. of radium bromide would evolve 0.5 c.c. of gas per day.

P. Curie<sup>4</sup> and Rutherford and Soddy<sup>5</sup> determined the rate of decay of the activity of the emanation, which was found to

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1. C. R. 132, 768 (1901).
  2. Berichte d. Chem. Ges. 35, 3605 (1902).
  3. Pro. R. Soc. 72, 204 (1903).
  4. C. R. 135, 857 (1902).
  5. Phil. Mag., April (1903).



be in accordance with an exponential law with the time, falling to one-half value in about four days. Curie<sup>1</sup> further determined that the rate of decay was not materially affected through such a wide range of temperatures as  $+450^{\circ}$  to  $-180^{\circ}$  C. Rutherford and Soddy<sup>2</sup> showed that the rate of decay for thorium emanations was practically the same at ordinary temperature as at that of liquid air.

Debiere<sup>3</sup> discovered the emanations of actinium. The loss of activity is most rapid, falling to one-half value in a few seconds. Giesel has obtained an intensely active emanation from the "emanating substance," which latter resembles lanthanum and cerium. By placing moist radium bromide on the screen, Giesel<sup>4</sup> noted the effect the radium emanation has upon a screen of phosphorescent zinc blende. With the slightest motion of the air the luminosity of the screen is observed to move in accordance with the air current. The same result could be observed by placing a small bit of the bromide in a tube and blowing air through the tube against different portions of the screen. Screens of barium-platinum cyanide and calcium sulphide did not become luminous under similar conditions. When the screen was charged with negative electricity the luminosity was most marked

The phenomenon upon which the spinthariscopes of Sir William Crookes<sup>5</sup> depends is based upon the bombardment of the zinc sulphide screen by the emanations. (Fig. 38.) Referring to this, Elster and Geitel<sup>6</sup> noted their previous observation of numbers of stars on an insulated zinc sulphide screen. A calcium tungstate screen showed only general or ordinary phosphorescence and no scintillations. Geitel<sup>7</sup> found the star-effect

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1. C. R. 136, 223 (1903).

2. Phil. Mag., May (1903).

3. C. R. 136, 146 (1903).

4. Berichte Chem. Ges. 35, 3608 (1902).

5. Chem. News 87, 241 (1903).

6. Phys. Zeit. May (1903).

7. Aus der Denkschrift der Kommission für luft-electrische Forschungen, München (1903), Chem. News 88, 29.

produced by soil emanations on a Sidot's screen charged negatively to 2000-3000 volts.

Crookes and Dewar learned that the scintillations ceased when the radium was cooled by liquid air, but the brilliancy was quite as marked when the Sidot's screen was cooled and the radium compounds were at normal temperatures. The highest vacuum attainable by cold does not affect the scintillations.

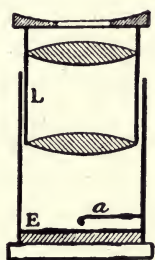


Fig. 38.

The Crookes Spinhtharoscope and the principle involved. On the end of wire pointer, *a*, is placed a tiny speck of a strong radium compound. This may be caused to move from place to place in front of a zinc-blende screen, *E*, from which it is less than a millimeter distant. Upon examining the screen in the dark, by means of the strong lens, *L*, which may be focused, one sees numerous beautiful scintillations, resembling the play of moonlight upon a rippling lake. The impression is given of the bombardment of the screen by many tiny particles, each flashing as it strikes the restraining phosphorescent substance.

Curie and Debierne<sup>1</sup> learned that in a vacuum a gas was given off from radium which produced excited activity on the glass walls of the vessel. The walls fluoresced, rapidly darkened, and affected the photographic plate. This gas did not show any lines in the spectrum, other than those of carbon dioxide, hydrogen and mercury. They, also, learned<sup>2</sup> that many substances were phosphorescent under the action of the

1. C. R. 132, 548 (1901).

2. C. R. 133, 931 (1901).

emanation. They found in general that substances which are phosphorescent in ordinary light become luminous, especially zinc sulphide. They also observed that phosphorescence was produced in Thuringian glass, showing most marked effects. Kunz and the author<sup>1</sup> noted that willemite is an even more sensitive detector of the  $\beta$ - and  $\gamma$ -rays than barium-platinum cyanide. Rutherford condensed the emanations of radium upon a crystal of that mineral with most brilliant fluorescent effects. The writer and Lockhart caused certain diamonds (tiffanyite) and minerals, as greenockite and wollastonite, to glow brilliantly when the emanations were condensed upon them. Pectolite and the spodumenes, especially the variety kunzite which responds to the  $\beta$ - and  $\gamma$ -rays, did not phosphoresce<sup>2</sup>. Soddy reports an observation contrary to this. The amount of excited activity deposited is proportional to the amount of emanation present, the distribution varying as the distance. Crookes<sup>3</sup> has caused white diamonds to assume the rare greenish color as a result of the action of the emanations. The jewels were buried for several weeks in radium bromide.

Rutherford and Soddy<sup>4</sup> measured the emanating power of different thorium compounds and learned that they varied very much, although the percentage of the thorium present in the compound was not very different. Rutherford<sup>5</sup> learned that the emanating power of ordinary thorium oxide is increased several times by heating the substance to a dull-red, but when heated to a white-heat the emanating power was greatly reduced. When the heat is maintained red, the emanation apparently continues to escape and the substance returns to its original value on cooling; whereas, after heating to a white-heat, on cooling it has only about ten per cent. of the original

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1. Science (N. S.), 18, 769 (1903).
  2. Science (N. S.), 18, 303 (1903).
  3. Chem. News 90, 1 (1904).
  4. Trans. Chem. Soc. 321 (1902).
  5. Phys. Zeit. 2, 429 (1901).

value. It has become "de-emanated." With lower temperatures, the emanating power of thorium decreases quite rapidly<sup>1</sup> being about ten per cent. of the original at the temperature of solid carbon dioxide. When the temperature is allowed to rise back to the ordinary, the original value is recovered. Rutherford and Soddy also verified the observations made by Dorn that the emanating powers of thorium and radium compounds are much affected by moisture, being greater in a moist than in a dry gas.

Thorium and radium compounds which have been de-emanated appear to recover the emanating property with time. It is not known whether this is due to a renewal or alteration of the substance which produced the emanation, or whether the intense heating simply changed the rate of escape of the emanation from the solid. The physical properties of thorium oxide are altered by intense ignition. According to Rutherford the color changes from white to pink and the oxide becomes denser<sup>2</sup> and is less soluble in acids. He dissolved a de-emanated oxide, precipitated it as hydroxide, and again converted it into the oxide. At the same time a sample of the ordinary thorium oxide was subjected to similar treatment. "The emanating power of both of these compounds was the same and was from two to three times greater than that of ordinary thorium." It should be noted that Rassignal and Gimmingham<sup>3</sup> have determined the rate of decay of the emanations as 51 and not 60 seconds, as given by Rutherford.

As a general rule, an increase of temperature in a solution of a salt of thorium or radium greatly increases the emanating power. From this it would seem that the original power of producing emanation persists in the atom. (See Chapter V.)

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1. Rutherford and Soddy, *Phil. Mag.*, Nov. (1902).

2. See the author's paper, "Thorium; Carolinium, Berzelium;" *Journ. Am. Chem. Soc.* 26, 922 (1904).

3. *Phil. Mag.* (6), 8, 107.

Henning<sup>1</sup> found that the radio-activity induced in metallic wires by thorium oxide depended upon the surface area of the wire, the volume of the containing tube, the fall of potential, and the thickness of the layer of the thorium oxide.

Rutherford and Soddy,<sup>2</sup> in a comparative study of the emanations of radium and thorium, found that as far as that property is concerned, they are closely allied, both producing radio-active emanations, and they in turn excite radio-activity in surrounding objects. The difference is very marked, however, in the rate at which the activity of the emanation decays. The intensity of the thorium emanation falls to one-half value in one minute, and that of the radium in about four days; while the excited radio-activity due to radium decays much more rapidly than that produced by thorium.

Curie and Danne<sup>3</sup> have shown that the radium emanation is absorbed by lead, paraffin and caoutchouc.

Touching the rate at which emanations are given off by solid radium compounds or when they are in solution, Rutherford and Soddy<sup>4</sup> have called attention to the following interesting point:

By theory, the amount of emanation stored up in a non-emanating radium compound is likely to be nearly 500,000 times the amount produced per second by the compound. By experiment the figure obtained was 463,000. Taking other things into consideration, this would indicate that the production of, or ability to produce, the emanation is the same with the solid compound as when in solution. It is occluded in the solid, and therefore given off slowly, while in a solution it is given off as fast as produced. Rutherford is of the opinion that the occlusion is not connected with the radio-activity of radium. The apparent occlusion of helium by minerals is analogous. The gas is driven out only in part by heat, com-

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1. Ann. Phys. (IV), 7, 562 (1902).
  2. Phil. Mag. (IV), 5, 445 (1903).
  3. Compt. Rend. 136, 364 (1903).
  4. Phil. Mag., April (1903).



pletely by solution. The writer and Lockart heated nearly all the rare-earth minerals and condensed the gases evolved by liquid air. All the helium bearing minerals gave off an emanation or something of a similar nature, which on refrigeration caused diamonds and a Sidot's blende screen to fluoresce. Other radio-active minerals, not containing helium, failed to respond in a like manner. Strutt<sup>1</sup> heated several minerals, samarskite, pitchblende, fergusonite, malacone, zircon, and monazite, collected the emanation evolved and measured the rate of decay. No new emanation was recognized. By drawing air over the cold minerals he learned that only a very small portion of the emanation is given out unless they be heated. All the minerals used contained helium and one, malacone, has been shown by Ramsay and Travers<sup>2</sup> to contain argon as well.

The question of the origin of the emanations of thorium, which according to Rutherford and Soddy<sup>3</sup> are produced by thorium-X and not thorium itself, present much that is of interest. Thorium oxide, freshly prepared by heating the hydroxide produced by precipitation with ammonia, shows no emanating power. In a month it has nearly reached a maximum in its recovery. The thorium-X, obtained by evaporating the filtrate from the ammonium hydroxide precipitation, gives out profuse emanations. The emanating power decreases rapidly and by the time the thorium has recovered its normal activity, the emanating power of the thorium-X has nearly disappeared. Considering the rates of decay and recovery, apparently the radiations are produced as thorium-X is changed into the emanation. With radium, however, no intermediate stage—radium-X—has been observed. Rutherford regarded the emanation as being produced directly from the element.

The emanations of radium and thorium were thought at one time to give rise only to  $\alpha$ -rays. Curie and Debierne<sup>4</sup> found

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1. Proc. Roy. Soc. 73, 191.

2. Proc. Roy. Soc. 64, 131.

3. Phil. Mag., Nov. (1902).

4. C. R. 133, 931 (1901).

the amount of excited activity in a closed vessel containing a radium compound unaffected by the pressure and nature of the gas. The rate of decay of the emanation has been shown to be the same under all conditions of concentration, pressure and temperature, provided the rate of supply of the emanation be constant. (Fig. 39.)

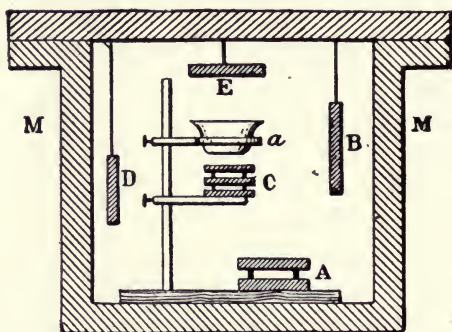


Fig. 39.

Rendering bodies active in a closed receptacle through the influence of the emanations. The radium salt is placed in a small dish, *a*, near various substances, *A*, *B*, *C*, *D* and *E*, it matters not what be their composition (lead, copper, glass, cardboard, ebonite, etc.). These substances acquire activity, may be removed, and their activity measured. The activity increases from the beginning according to the time which the substance remains in the receptacle. A limit-value is reached after a certain length of exposure.

#### *Chemical Nature of the Emanations.*

In their earliest experiments Rutherford and Soddy<sup>1</sup> submitted thorium emanations, obtained by passing air over thorium oxide, to a most stringent treatment. They found it unaltered after being heated by electricity to the highest temperature attainable, when passed over platinum black, cold and hot, red hot lead chromate, magnesium powder, and zinc dust. The

1. Phil. Mag., Nov. (1902).

only gases known to withstand such drastic treatment are those of the argon family.

Later Rutherford and Soddy<sup>1</sup> sparked the emanation from radium in a glass tube, containing oxygen and an alkali, heated it red hot in a magnesia lime tube for several hours, and observed no diminution in its rate of discharge. They learned that a tube containing a large amount of radium emanations phosphoresced brightly under the influence of the rays given out. The removal of the emanations from one point to another in the tube was easily observed in a darkened room by the luminosity of the glass. The luminosity of the emanation increased when the gas was compressed.

Rutherford and Brooks<sup>2</sup> and Curie and Danne,<sup>3</sup> as reverted to, learned that the emanation of radium, like a gas, always divided itself between two connected reservoirs in proportion to their volumes. By determining the co-efficient of the diffusion of the emanation in the air, they learned that the molecular weight of the gas must be large. The same was demonstrated for thorium. Crookes<sup>4</sup> directed attention to the difference in the rate of diffusion of the "radiant matter" of radium, actinium, and polonium through air. The last is the slowest. The "emanations" from hydrogen peroxide are not carried through a tube by air.

Wallstade,<sup>5</sup> by determining the co-efficient of the diffusion of radium emanations into various liquids, arrived at the same conclusion as to the gaseous nature of the emanations.

Rutherford and Soddy<sup>6</sup> learned that the emanations from thorium and radium were condensed at a temperature of liquid air. By placing a phosphorescent zinc sulphide screen, or a small piece of willemite in a tube, the presence of the emanations is readily observed through the glowing of either of these

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1. Proc. Roy. Soc. 72, 204 (1903).

2. Chem. News (1902).

3. C. R. 136, 1314 (1903).

4. Proc. Roy. Soc. 69, 413 (1902).

5. Phys. Zeit. 4, 721 (1903).

6. Phil. Mag., Nov. (1902), and (6), 561.

substances. The luminosity of the screen is due in part to the radiation from the emanation and in part to the excited radiation caused by it. The accompanying figure (Fig. 40) illustrates the very simple methods for the condensation of the emanations. The temperature of the condensation of the thorium emanation is not sharply defined, but is probably  $-120^{\circ}$  C, while the temperature for the radium emanation is about  $-150^{\circ}$  C. Their actual quantity is almost infinitesimally small. They are invisible and unrecognizable, but their presence is readily detected by their property of radio-activity.

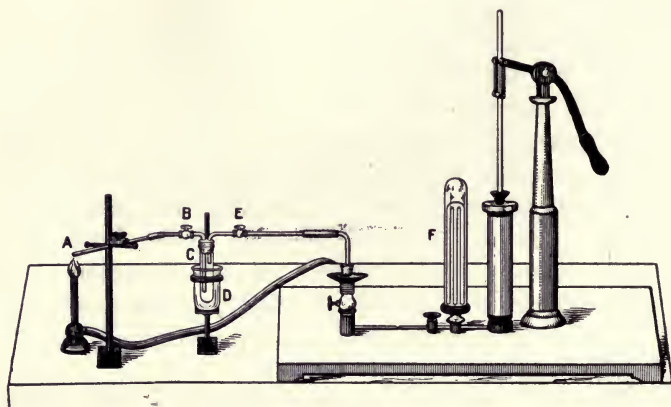


Fig. 40.

Simple apparatus for condensing the emanations. A radium compound or other radio-active substance giving emanations is placed in small tube, A, connected by heavy rubber tubing to another tube, which projects through a two-hole rubber stopper nearly to the bottom of the thick-walled test-tube (C). The exit tube, also provided with a cock, passes from just within the test-tube through the stopper to the vacuum pump. Within the test-tube may be placed a small piece of Willemite (Kunz and Baskerville), a diamond, or zinc sulphide screen. The tube is exhausted by opening cock E, B remaining closed. After exhaustion E is closed, C placed in liquid air in the Dewar bulb (D). A is gently heated, B opened and at once the emanation rushes over and is condensed on the materials mentioned. The glow is very beautiful. By closing B the imprisoned emanation may be held for hours or days.



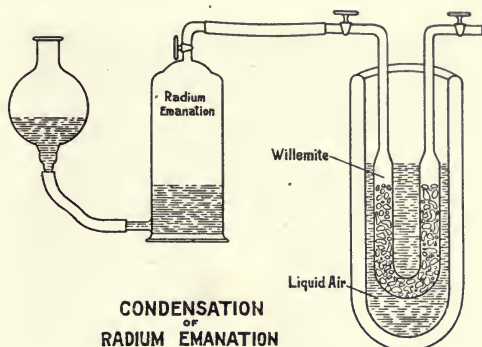


Fig. 40 A.

The emanations from thorium and from radium are quite distinct from each other in two particulars: first, the difference in condensation, as just noted; and second, their difference in radio-activity. The rate of decay of the radium emanation is about 5000 times slower than that of the thorium emanation.

As a result of the investigation of the heat emission of the radium emanation, it has been learned by Rutherford and Barnes<sup>1</sup> that the emission corresponds, approximately, to the activity as measured by the  $\alpha$ -rays. That is, it accompanies the expulsion of the  $\alpha$ -particles and is proportional to the number expelled. The emanation is responsible for about seventy per cent. of the heat effect of radium. The amount of the emanation is extremely small, but it has been calculated by Rutherford that one c.c. of the emanation at standard pressure and temperature would emit about  $3 \times 10^7$  gram calories of heat. This would indicate that heat would be produced at such a rate as to melt an ordinary glass tube which might be used to contain the emanation in quantity.

"If the atomic weight of the emanation is taken to be about 200, it can be calculated that one pound weight of the emanation would initially radiate heat at the rate of about 8000 horse-power, and in the whole course of its heat emission would radiate an amount of energy corresponding to 40,000 horse-power

1. Phil. Mag. (6), 207 (1904).



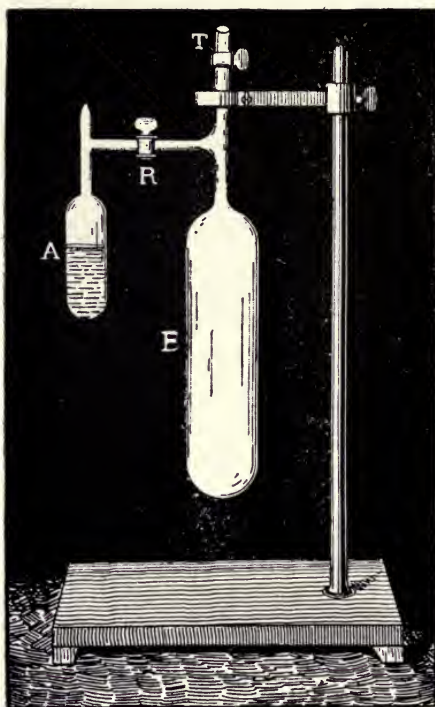


Fig. 41.—PHOSPHORESCENCE CAUSED BY THE EMANATION OF RADIUM.

A vacuum is formed in this reservoir through the tube, T, and air charged with emanation is afterward let in from a reservoir, A. The tube, A, contains a solution of a radium salt, and the emanation disengaged has accumulated in the gaseous part. As soon as the cock, R, is opened, the reservoir, B, becomes very luminous, and the light emitted by the sulphide of zinc is sufficiently bright to permit of reading being done at a distance of 4 or 8 inches from the tube.

days. In order to obtain such an amount of emanation about seventy tons of radium would be required.”

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1. “Radio-activity,” Rutherford, p. 247, MacM. Co.

Beilby<sup>1</sup> found that glass was decomposed in the neighborhood of certain hot metals, as gold and platinum. This intensification of chemical action he attributed to the emanations from the metals, although they are not radio-active, as the term is used.

Sir William and Lady Huggins<sup>2</sup> photographed the phosphorescent spectrum of a radium compound by a 72-hour exposure. They are reported<sup>3</sup> as having found five of the eight lines observed as coincident with helium. This appears to have been erroneous, as the lines were really found to have agreed in position and intensity with the band spectrum of nitrogen. Later Crookes and Dewar<sup>4</sup> learned that the nitrogen spectrum did not appear when the radium bromide was placed in a highly exhausted quartz tube. Yet Curie and Dewar<sup>5</sup> secured notable amounts of nitrogen which was occluded by the purest radium bromide. .4 of a gram of pure dry radium bromide were left three months in a glass bulb connected with a small Geissler tube in a mercury manometer, a high vacuum being made in the whole apparatus at the beginning. During the entire three months one cubic centimeter of gas per month at atmospheric pressure was given off continuously from the radium salt. Spectroscopic examination showed only the presence of hydrogen and mercury vapor, the former doubtless due to a small amount of water, native with the radium salt and decomposed by the radium. The same sample was taken to England and used by Dewar at the Royal Institution for measuring the heat given off at low temperatures. It was in a quartz bulb provided with a tube of the same substance. The bulb was evacuated and the tube heated to the fusion point of salt. The gas given off by the bromide was collected by a mercury pump. After passing through a set of new tubes, cooled by liquid air, which

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1. British Association, Southport Meet. (1903), Chem. News 88, 178.

2. Proc. Roy. Soc. 72, 196 and 409 (1903).

3. Science (N. S.), 18, 186 (1903).

4. Brit. Assoc. (1903).

5. Compt. Rend. 138, 190.

condensed the greater part, the remainder of the gas was collected in a test tube over mercury. This amounted to 2.6 c.c. at atmospheric pressure. Part of the radium emanations was brought over and was radio-active and luminous. The light given off by the gases in the test tube, after three days' exposure with a photographic quartz spectroscope, gave a discontinuous spectrum with three lines coinciding with the three principal bands of nitrogen, namely, 3,800, 3,580, and 3,370. The glass tube took on a deep violet hue and half the volume of gas was absorbed during the three days. On passing a spark through the gas in a Geissler tube, the nitrogen bands also appeared. On condensing nitrogen with liquid hydrogen, a high vacuum was produced in the Geissler tube and the spark showed only the nitrogen present. The quartz tube was heated until the bromide of radium melted and deprived of all the occluded gases, sealed by an oxy-hydrogen blow-pipe, when the vacuum was made, and carried to Paris. Twenty days after the sealing Deslandres examined it spectroscopically by illuminating the tube with an induction coil, using two rings of tin foil around the tube as the poles and secured the *entire spectrum of helium*. This was noted even after an exposure of three hours with the quartz spectroscope.

These observations were in accord with the analogous investigations of Ramsay and Soddy<sup>1</sup>.

Rutherford and Soddy<sup>2</sup> first suggested that the emanation might consist of helium. The latter carried the problem to the master Ramsay, whose laboratory possessed superb facilities for handling and investigating small amounts of gases. They removed the hydrogen and oxygen, liberated in large quantities from a water solution of the bromide, condensed the emanation and carbon dioxide by liquid air and found the characteristic D<sub>3</sub> line of helium. Later they found not only the complete spectrum of helium  $\lambda\lambda$ - 6677, 5876, 5016, 4972, 4713 and 4472, but three other lines which were not identified, namely,  $\lambda\lambda$ - 6180, 5695, and 5455.

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1. Compt. Rend. 138, 190.

2. Nature 246 (1903), and Proc. Roy. Soc. 72, 204.

For a fuller discussion of the emanations see Chapter V.

*Excited Radio-Activity.*

Substances in contact with radio-active bodies acquire the power of affecting a photographic plate, and ionizing gases. In making certain radio-active experiments, therefore, it is of the utmost importance that precautions be taken to avoid the presence of other radio-active substances in the room.

The Curies<sup>1</sup> first observed this property of inducing activity by radium and Rutherford<sup>2</sup> noted it for thorium. Solid substances placed within a closed vessel, which contains an emanating compound, become radio-active. The intensity of the radio-activity varies directly with the proximity. With radium preparations it is different. After an exposure of several hours the excited activity is independent of the position or composition of the plates. Mica, ebonite, cardboard, and copper exhibit equal amounts of activity. It is dependent upon the extent of surface exposed. (See Fig. 39.)

Becquerel<sup>3</sup> examined the secondary radio-activity of metals, which was attributed to the absorption of the incident radiation. The phenomenon appeared to correspond to that of fluorescence or phosphorescence with regard to light and analogous to the secondary rays derived from Röntgen rays discovered by Saginac. They are less penetrating than the original, but consist of portions, (a) not deviable by a magnetic field, but easily absorbed, (b) deviable and apparently identical with cathode rays, and (c) not deviable, but very penetrating.<sup>4</sup>

This property of exciting radio-activity appears to be due to the emanations and proportional solely to the amount present. It may be concentrated upon the negative electrode in a powerful electric field.

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1. C. R. 129, 714 (1899).
  2. Phil. Mag., Jan. and Feb. (1900).
  3. Compt. Rend. 132, 7.
  4. Compt. Rend. 132, 7, 12, 371 (1901).



Fig. 42.

Residual activity. The pen was radiographed by a glass tube, which contained 5 mgms. of radium bromide, but which had been empty a month.

The activity which platinum wire acquires by being placed in thorium solutions may be removed by acids like nitric, hydrochloric and sulphuric.<sup>1</sup> The deposited radio-active matter may be largely removed by scrubbing the wire with emery paper. No increase in the weight of the wire has been observed before or after it becomes active. No difference is noted under the microscope. Whatever it is, therefore, it must be vastly more active than radium itself. Rutherford has termed this "radio-active matter" Emanation-X, as it is quite distinct chemically and physically from the emanation which produces it. This we appreciate at once when we recall that the emanation is a gas, unaffected by chemicals, while the emanation-X is a solid and readily soluble in acids. Platinum wire becomes active by exposure to the emanations of thorium, but loses its activity when raised to a white-heat. Gates<sup>2</sup> learned that the activity was not destroyed, however, but transferred to the walls

1. Rutherford, *Phil. Mag.*, Feb. (1900).

2. *Phys. Review*, p. 300 (1903).



of the vessel in which the heating had taken place. The same was learned of activity induced by radium.

Miss Brooks has shown that dust particles, as other solids, acquire radio-activity when enclosed in the presence of emanations. M. and Mme. Curie<sup>1</sup> found that substances after a long exposure to radium did not lose all of their acquired activity. Giesel<sup>2</sup> has found that the radiations from an excited platinum wire consist entirely of  $\alpha$ -rays. Rutherford<sup>3</sup> found the residue from a solution of the deposited matter after evaporation retained its activity. It was retained when it was enveloped in a copper coating electrolytically deposited. Von Lerch<sup>4</sup> studied the emanation-X of thorium and learned that copper or magnesium wires served to take up most of the active matter. When these metals were dissolved and precipitated as different compounds the activity remained and decayed at the normal rate; namely, one-half within 11 hours.



Fig. 43.

Induced radio-activity. The key was radiographed with water rendered active by allowing a tube of radium salt to remain in it some time and then removing it.

Barium sulphate also carried down the emanation-X by precipitation. Different metals dipped into active solutions varied in their conduct. Zinc removed almost all of the activity. Iron, nickel, aluminum, copper, lead and cadmium, also, became active, while platinum, palladium and silver did not. Pegram<sup>5</sup>

1. Thesis, 1903, 116.

2. Ber. d. deutsch. Chem. Ges. 36, 2368 (1903).

3. Rutherford, Phys. Zeit. 3, 254 (1902).

4. Annal. (4), 745 (1903).

5. Phys. Review, Dec. (1903).

electrolyzed thorium solutions obtaining a radio-active deposit of lead peroxide on the anode from the commercial and the



Fig. 43.

This is a radiograph of a gold fish which had been placed in water rendered radio-active by having suspended in it for twenty-four hours a closed tube containing ten milligrams of radium of high activity. By this process the water was rendered radio-active and the fish was then placed in the water, and although the radium had been entirely removed, the fish itself was rendered radio-active, and when placed on a photographic plate, photographed itself by its own radio-activity.



Fig. 44.

Induced radio-activity. The fish made the auto-photograph after being subjected to the action of radium bromide, 300,000 activity.

so-called chemically pure salts. With pure preparations furnished by the author no visible deposit was found, but the anode was active. Its activity decayed to half value in an hour, whereas the rate was 11 hours for the commercial preparation, the normal rate determined for all the thorium preparations hitherto used. This emphasizes a point to which the author has frequently directed attention; namely, that most investigations on the activity of thorium have not been made with preparations of sufficient purity. Nor have preparations been used whose life-history has been known.

Rutherford and Barnes<sup>1</sup> determined the heating effect of radium emanations. This can best be illustrated, as shown by Rutherford:

Active products	Nature of rays	Percentage proportion of total activity measured by rays	Percentage proportion of total heating effect
Radium freed from active products	$\alpha$ rays	25	25
Emanation	$\alpha$ rays	18	41
Emanation X (1st change)	$\alpha$ rays	15	
“ (2d change)	No $\alpha$ rays	0	34
“ (3d change)	$\alpha, \beta$ & $\gamma$ rays	42	

The heating effect which accompanies the expulsion of the  $\alpha$ - particles appears to be approximately proportional to the number expelled.

Rutherford<sup>2</sup> learned that under low pressures the excited activity produced by thorium is found on both anode and cathode, it matters not what the strength of the electric field may be.

1. Phil. Mag. (VI), 7, 202 (1904).

2. Phil. Mag., Feb. (1900).

Curie and Debierne<sup>1</sup> learned that the amount of excited radio-activity produced by a radium compound was much reduced when the gas within the vessel was kept at a low pressure. They<sup>2</sup> also learned that induced activity would result from the presence of a solution of a radium salt. The action is more regular and intense when a solution is used. It produces phosphorescence of glass. This is independent of the position of the radium solution provided sufficient time be allowed.

The excited radio-activity is attracted to the cathode in a strong electric field. Fehrle<sup>3</sup> learned that excited activity followed the lines of force in an electric field. It appears that the radio-active matter, therefore, is transported by positively charged carriers.

Giesel<sup>4</sup> reports that his "emanating substance" gives rise to a type of radiations which he termed E-rays. By electrifying a zinc sulphide screen negatively, more brilliant luminous effects were produced, which would indicate that the carriers of the excited activity of his emanation substance have a positive charge. Batelli and Maccarone,<sup>5</sup> by using an especially sensitive electrometer of small capacity, suitable for working at ordinary or liquid air temperatures, found that the emanation carries no charge. They are not atomic residues which the positive ions have lost, but the positive ions themselves. McClelland,<sup>6</sup> also, arrived at the same conclusion.

Debierne<sup>7</sup> showed that barium could be rendered artificially active by precipitation from a solution of actinium. A very active barium chloride was made in this way, concentrated like radiferous chloride, but no spectroscopic lines of radium were

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1. C. R. 132, 768 (1901).

2. C. R. 133, 23, 931.

3. Phys. Zeit. 3, 130 (1903).

4. Berichte 36, 342 (1903).

5. Atti. R. Acad. d. Lincei Roma (5), 13, 539 (1904).

6. Phil. Mag. 6, 355 (1904).

7. C. R. 131, 137 (1900).

obtained. The activity of the barium decayed to about one-third its value within three months.

Debierne,<sup>1</sup> also, obtained a large amount of emanation from actinium. Its activity decays very rapidly. The emanation produced excited activity on adjacent bodies. He attributed the excited activity of actinium to "ions activants."

The induction of radio-activity and ionization of gas are quite distinct from one another. Therefore, the actinium emanation must be regarded as containing two sorts of energy.<sup>2</sup> That is, the substance containing actinium seems to emit a second emanation which decays much more slowly than the first one described.

Giesel, four years ago, found that a stick of bismuth would become active when placed in a radium solution. He intimated that polonium was practically radiferous bismuth. Madame Curie repeated the work by fractionation and obtained a bismuth two thousand times as active as uranium.

Again, Giesel<sup>3</sup> found that his bismuth plate remained active after every effort had been made to remove all traces of radium. It gave out only  $\alpha$ -rays and therein resembled polonium and radio-tellurium.

Mme. Curie<sup>4</sup> developed a law for the dissipation of excited radio-activity in an unconfined air space. The intensity for radium is reduced to one-half value in twenty-eight minutes. For actinium and thorium the loss requires greater time. Within a closed space, the emanation from radium may be said to disappear spontaneously as a function of the time, going to one-half in four days, according to Rutherford.

According to the law for unconfined spaces, the activity induced should be almost imperceptible. Certain substances, as celluloid, paraffine, and caoutchouc, however, lose their acquired activity with great slowness, sometimes requiring fifteen or

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1. C. R. 136, 446, 671 (1903).

2. C. R. 138, 411 (1904).

3. Berichte 36, 2368 (1903).

4. Thesis (1903).



more days. In losing it, they also induce radio-activity. Doubtless, this lag, or special induced activity, has much to do with the unique observations of Metzenbaum,<sup>1</sup> who caused zirconium and yttrium compounds to affect a sensitive photographic plate similarly to thorium compounds. His experience is unique and contrary to that reported by others. Perhaps it may be due to the admixture of small amounts of radium. This is the explanation he offers for the activity of thorium. Haitinger has extracted radium from commercial thorium oxide.

Metzenbaum<sup>2</sup> produced skiagraphs with metallic aluminum by placing it directly on the sensitive gelatine. A shield of black paper or glass prevented the darkening of the plate. This can be readily attributed to chemical or electro-chemical action. He placed closed tubes of radium preparations in various powders for several days. After removal, he obtained negative results as to induced activity, tested photographically and with the electroscope.

Previously, much prominence was given by the secular press to the reports of exciting activity in salt and other solutions, which might be used internally for specific therapeutic effects. (See Chapter VI.)

Heydweiller<sup>3</sup> reported no loss of weight from a closed radium tube. Dorn,<sup>4</sup> however, reported diminution, while Forch observed no change. Davis, in our laboratory, was unable to detect any loss in weight of a closed tube containing a gram of chloride, 7000 activity. So far no satisfactory experimental evidence, as to the loss of weight by radium compounds, has been offered. Piffard<sup>5</sup> calls attention to the fact that no authoritative statement has been given as to the rendering of

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1. "Radium, Radio-active Substances and Aluminum," Cleveland, O., '04. *Scientific American*, May 14, 1904, and *Cleveland Med. J.*, May, '04.

2. *Loc. cit.*

3. *Phys. Zeit.* 4, 81 (1902).

4. *Phys. Zeit.* 4, 530 (1903).

5. "A Few Words Concerning Radium," *Medical Record*.

water or other substances radio-active by the presence of a closed tube of radium. He further detected defects in tubes, air bubbles, etc., and regards the statements concerning induced activity by means of closed tubes as based upon the use of defective tubes. As Curie and Rutherford have shown, induced activity requires a naked exposure of radio-active bodies. It is superficial, the real extent depending entirely upon the depth to which the emanations or their products have penetrated. It has been definitely proved that the emanations consist of material particles. Their expulsion and consequent transference, when radio-active substances are exposed, must, therefore, mean a corresponding loss in weight of the original substance.

To explain the phenomenon of induced radio-activity, two hypotheses have been put forward. The first states that the inactive molecules of almost any substance after being mixed with an active substance like radium temporarily acquire the property of radio-activity. The second hypothesis states that inactive bodies become active by association with active substances by removing a small portion of the latter, or by removing a radio-active product of the element. Thus the excited activity may be permanent or temporary, decaying according to the law governing the radio-active product removed. From the work of von Lerch, already mentioned, the latter appears to be the most acceptable explanation.

## CHAPTER V.

## THEORIES OF RADIO-ACTIVE PHENOMENA.

Naturally the ideas of Madame Curie, which led to the brilliant discovery, deserve first mention. Her experimental data warranted the assumption that radio-activity is an atomic and not a molecular phenomenon, although she does not commit herself unreservedly to that explanation.

Becquerel<sup>1</sup> used the following hypothesis for his investigations: radio-active matter, according to J. J. Thomson, consists of negatively and positively charged particles. As shown by Thomson's work, the negative particles have a mass of about 1/1000 that of hydrogen, and the positive particles have a mass about equal to hydrogen. The former (or  $\beta$ -rays) are projected at a very high velocity, while the latter are comparatively sluggish, constituting the emanation which may be deposited upon the surface of bodies and which give rise to excited activity.

Becquerel,<sup>2</sup> further, having noted that the activity of uranium was not constant, as previously noted by Giesel and Crookes, suggested that the emission of the deviable rays be identical with the cathode rays and the cause of the non-deviable radiation so much like the X-rays. It was thus comparable to the evaporation of an odorous body. The dissipated energy would be given out from the active body itself, but the corresponding loss of weight would be too small to be observed.

Rutherford and McClung<sup>3</sup> previously learned that the energy given out in the form of ionizing rays was 3000 gram-calories per year in radium, 100,000 activity, or with the pure

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1. C. R. 133, 979 (1901).

2. C. R. 133, Dec. 9 (1901).

3. Phil. Trans. 25 (1901).

radium preparation, 1,500,000 activity, an emission of energy in the gas, as  $\alpha$ -rays, of about 45,000 gram-calories per year. It was suggested that this energy might be derived from a re-grouping of the atomic constituents of radio-active elements. Even before that, Rutherford<sup>1</sup> believed that thorium emanations and excited activity were due to radio-active matters. With Brookes, and later Soddy, he learned that the emanations of thorium and radium behaved like gases, that they produced excited radio-activity, that they diffused through air like gases of heavy molecular weight, and that they behaved very much like the chemical inert gases, with the exception that they were dissolved in some acids and not in others.

Curie<sup>2</sup> differed from Rutherford, calling attention to the fact that no spectroscopic evidence of the gas had been obtained and, that, also, the emanation disappeared when in a sealed vessel. He regarded the emanation as consisting of centres of condensed energy, attached to gas molecules, and moving with them. Rutherford<sup>3</sup> claimed that the failure to detect the gas spectroscopically could be accounted for through the minute quantity of the emanation present (one gram of radium produces  $3.3 \times 10^{-4}$  c. c. at atmosphere pressure and temperature<sup>4</sup>) although the electrical and phosphorescent actions were very marked with the amounts to be had. Rutherford and Soddy<sup>5</sup> studied uranium, thorium and radium, condensed the radio-active emanations at the temperature of liquid air, demonstrated that the  $\alpha$ -rays consisted of positively charged bodies, atomic in size, and projected with a great velocity. This proof of the materiality of the emanations forced upon them the necessity for assuming the continuous production, by thorium and radium, of new kinds of active matter which possess temporary activity and differ chemically from either of those two

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1. Phil. Mag., Jan. and Feb. (1900).

2. C. R. 136, 223 (1903).

3. Phil. Mag., April (1893).

4. Phil. Mag., May (1903).

5. Trans. Chem. Soc. 81, 321, 837 (1902), and Phil. Mag., Sept. and Nov. (1902), Feb., Apr. and May (1903).

elements. Further, it was learned that the radio-activity assumes a constant, being a resultant equilibrium between the processes of production of active matter and the alteration of those already produced.

Curie and Laborde<sup>1</sup> suggested that the heat may as well be supposed to come from the breaking up of the radium atom as from energy absorbed by it from some outside source. J. J. Thomson<sup>2</sup> postulated the emission of energy as being due to some internal changes in the atom, and that a large store of energy would be released by a contraction of the atom.

Fillipo Re<sup>3</sup> put forward his belief that particles have previously been free and that they constitute nebulous formations of extreme tenuity. In time they became reunited around centres of condensation, giving rise to small suns, as it were, which by ulterior contraction, take stable and definite form. These make up the atoms of ordinary chemical elements. As we know them, they may be compared to small extinct suns. The larger suns, not yet extinct or cold, constitute the atoms of radio-active bodies, hence the heat absorbed with these substances. Latterly this idea has come forward with greater prominence.

Hudson Maxim<sup>4</sup> accounted for its luminous, heat-giving effect by asserting that radium has a property opposite to ultra-violet rays, that the high electric waves impinging upon radio-active substances slow down to waves of lower pitch, some corresponding with visible light, others with heat. In the same manner an opaque body like a piece of smoky glass, will get hot in the direct sunlight by a slowing down of the higher light rays to the lower pitch, which are sensed as heat. Two years later Lord Kelvin, in a paper before the British Association, presented the same theory, using nearly identical illustrations.

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1. C. R. 136, 673 (1903).
  2. Nature (1903), 601.
  3. C. R. 136, 1393 (June 8, 1903).
  4. Electrical Age (1901).



DuPont<sup>1</sup> suggests that radio-active substances are catalytic agents, radium being very powerful; that radio-activity is a form of catalysis, and that the action which radium has upon surrounding bodies is due to this cause.

A catalytic agent is a body which by its mere presence accelerates chemical reaction or causes chemical reactions to take place within other bodies which would not react upon each other or would react very slowly, except for its presence. As an illustration, we may cite the action of platinum in the formation of sulphur trioxide from sulphur dioxide and oxygen.

The converse order was suggested, namely, that radio-activity might be used as a key to the solution of the problem of catalytic action. Radium has the effect of discharging electrically charged bodies. Riecke regards atoms as electrically charged. Perhaps the effects of radium upon animal tissue may be due to the discharging of negative electricity, which holds certain molecules from uniting with other molecules, thereby bringing about chemical reactions which under normal conditions are impossible of being effected.

Attention is called to the phenomena observed within a spinthariscopes. The emanations do not resemble light rays thrown off from luminescent bodies, but are more like a meteoric shower, or a lot of miniature bomb shells exploding. Maxim likens the action of radium to the familiar theory of the thunder storm. That is, small aqueous vesicles forming the clouds, each vesicle charged with a small amount of electricity, unite with one another forming larger vesicles. As they are spherical the larger vesicles show a smaller surface in proportion to the mass; consequently the electrical tension upon the surfaces becomes greater as the vesicles grow into drops of water and it is the uniting into one great electrical spark of an infinite number of small electrical sparks passing from drop to drop that produces the lightning flash and clap

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1. Scientific American Supplement, Apr. 9 (1904), p. 23631.

of thunder. Radium, acting upon the atmosphere in contact with it, or in its immediate vicinity, discharges the electricity from certain molecules to certain other molecules, producing miniature reactions. Possibly these miniature electrical discharges produce light; that is, one of these tiny flashes of lightning. Were our ears acute enough it might be possible to distinguish these infinitely small claps of thunder.

The production of helium from radium is attributable not to the conversion of any portion of the radium into helium, but to the production of helium from the atmosphere or other medium by the catalytic action of the radium. Therefore, the energy does not come from the radium, but exists in the atmosphere as potential energy and is allowed by the radium to become kinetic energy, just as the platinum causes sulphur dioxide and oxygen at certain temperatures to combine, producing heat.

From the observations made, Rutherford and Soddy<sup>1</sup> suggested that helium might be the production of the disintegration of the radio-active elements.

Gutton reported observations which, perhaps, have a bearing upon the theory of radio-activity. He found that when the lines of force of magnets are not parallel that luminous effects may be produced upon a phosphorescent screen. de Hemptinne,<sup>2</sup> however, was unable to verify the observations.

Concerning the radio-activity of thorium, Baskerville<sup>3</sup> has called attention to the dividing of thorium into constituents which differ in their activity. Hofmann and Zerban<sup>4</sup> call attention to the important fact, from a chemical point of view, that thorium, which is radio-active, comes from minerals containing uranium. All the thorium preparations, with which physicists and chemists have usually worked, have, as a rule, come from complex minerals which, probably, contained variable amounts of uranium. The above mentioned workers

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1. *Phil. Mag.* (1902), 582; (1903), 453, 579.

2. *C. R.* 138, 754.

3. *J. A. C. S.* 26, 922.

4. *Berichte* 35, 531, and 36, 3093.

extracted from certain minerals, Norwegian gadolinite, yttrio-titanite, and orthite free from uranium, thorium which did not possess any radio-activity. Haitinger<sup>1</sup> has succeeded in extracting radium from thorium, which was prepared from Brazilian monazite sands. The writer and Zerban have later obtained a thorium preparation from a South American mineral, which is absolutely inactive.

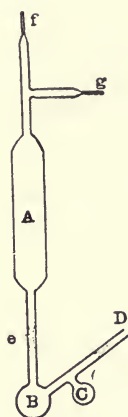


Fig. 45

Boltwood's apparatus for showing the ratio of radium to uranium in minerals. A weighed quantity of powdered mineral is placed in bulb B. The acid to be used for decomposing the mineral is placed in C. After decomposition, which is brought about by inclining the tube so the acid may come into contact with the mineral, the apparatus is allowed to stand a few days until equilibrium is reached. The tube A is then sealed off at e. The air and radium emanation are then removed from A by suction, introduced in an electroscope and the ionizing power determined.

Boltwood<sup>2</sup> concluded from a determination of the amount of radium in radio-active ore, and the rate of leakage of the electroscope, that the amount of radium present stands in direct proportion to the percentage of uranium.

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1. Haitinger and Peters, Sitzungs Berichte (Wien) 113, May, 1904.  
2. Eng. and Min. Journ. 77, 756.

It has been suggested by J. J. Thomson and Rutherford as very probable that radium is formed by the breaking down of the uranium atom. A final state of equilibrium and definite proportion between uranium and radium present in minerals was to be expected, which fact prompted Rutherford and Soddy<sup>1</sup> to suggest a complete study of the natural minerals. The improbability of any radium ore being found containing a greater portion of radium than pitchblende, because it contains the highest percentage of uranium, suggests itself.

Preliminary experiments on the relative amounts of polonium present in two different uranium minerals, showed by comparison that in all probability this element also varies directly with the percentage of uranium present.

Mendeleeff<sup>2</sup> insists that radio-activity indicates a material emanation. The arrival and departure of atoms are accompanied by disturbances which indicate waves of light.

M. and Mme. Curie<sup>3</sup> have presented a general theory concerning radio-activity as follows: It is an atomic property. Each atom acts as a constant source of emission of energy, which may be derived directly from the potential energy of the atom, or the atom may serve as a means whereby the energy may be borrowed from the surrounding air. Crookes<sup>4</sup> suggested that radio-active elements possess the property of abstracting energy from a gas. That is, in order to account for the large emission of heat from radium noted by Curie and Laborde, the moving materials might strike a substance and be released with a changed lower velocity, a production of heat resulting.

Lord Kelvin,<sup>5</sup> also, suggested that radium perhaps obtains its energy from an external source. It would be interesting

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1. Phil. Mag. 576.

2. "A Chemical Conception of the Ether," Longmans, Green & Co., '04.

3. C. R. 134, 85, 1902.

4. C. R. 128, 176 (1899).

5. British Assoc., 1903 Meeting.

to obtain inactive thorium and keep it for months in a vacuum and note whether or not the de-emanated body re-acquired its radio-activity.

Many are not as yet ready to accept the materiality of the cathode or  $\beta$ -rays. That the emanations are composed of definite particles is proved beyond question. Most active products emit only  $\alpha$ -rays, or at least they constitute by far the major portion of the radiations. No substance has yet been obtained and known for any length of time which gives out only  $\beta$ - or  $\gamma$ -rays, either alone or together. Rutherford states that the  $\beta$ - and  $\gamma$ -rays in most cases appear only in the last stages of radio-active processes. This statement must be modified in view of the work reported by Rutherford at the Congress of Arts and Sciences, St. Louis, 1904. (See end of chapter.) Perhaps then in time those bodies which emit only  $\alpha$ -rays may yield the other two forms of recognized energy. Such falls in well with the theory proposed by the writer<sup>1</sup> and Lockhart, which follows:

The elements of high atomic weight are electro-positive. The emanation particles bearing a positive charge are repelled. As they are lighter and gaseous, they are thrown away from the ponderous solids at a high velocity, about 1/10 that of light. These particles provoke an opposite charge producing ethereal stresses, cathode or  $\beta$ -rays. These acting upon any solid substance produce the  $\gamma$ -rays in the same manner that the Röntgen rays are produced without the Crookes tube through the influence of the cathode rays within. This appears to negate entirely the postulates of Crookes and J. J. Thomson as to the materiality of the cathode rays. It is maintained by some that while the presence of material particles may be accepted, they are the remnants of gas not removed in the exhaustion of the tubes. In fact the efficient modern Crookes tubes for the production of X-rays are arranged to keep a variable amount of gas present within. The gas particles serve as carriers of the

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1. "The Cause of Radio-Activity," Washington Section A. C. S., April 6, 1904.



negative charge and the assumption of the materiality of the cathode or  $\beta$ -rays becomes unnecessary.

J. A. McLennan<sup>1</sup> stated that the emanation from radium is not charged electrically. The radium atom gives off positively charged particles. "The emanation cannot be what remains of the atom after the emission of these rays, as it would then be negatively charged. The atom must have, therefore, parted with an equal negative charge, either by the emission of negative particles or in some other way."

Schenck<sup>2</sup> proposed a theory for radio-activity based on the hypothesis of electrons in phenomena of chemical equilibrium and more particularly in that one between oxygen and ozone which is controlled by the laws of mass effects.

Richartz has shown that ozone belongs to the group of radio-active substances and on being dissociated will become a conductor of electricity. In short, it would be converted into oxygen while giving off gaseous ions. On the other hand, its formation takes place whenever in certain electric phenomena gaseous ions are present and a reversible process analogous to the dissociation phenomenon occurs. If gaseous ions be considered as material particles, the ozone may be regarded as a chemical compound of electrons and oxygen, or an "electronide" of oxygen. Both electrons of atomical ions would be controlled by the mass law in the same way as electrolytic ions and electrical and neutral molecules. The hypothesis is suggested that radium and analogous substances might also be "electronides." The process might be analogous to the dissociation of calcium carbonate into calcium oxide and carbon dioxide. Probably radio-active substances should be produced by volcanic phenomena, as they are attended by violent evolution of electricity. In many slow reactions giving rise to the formation of ozone, the presence of gaseous ions has lately been ascertained. It is probable that many, if not all, reactions are attended with the presence of such gaseous ions in

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1. Phil. Mag. 6, 7, 355 (1904).

2. Pruss. Acad. of Science (1904).

variable quantities. On the other hand, hydrogen dioxide is analogous to ozone, giving off so-called emanations which do not influence photographic plates through a sheet of aluminum. It should equally be considered as an electronide. In order to produce a luminous sensation on the eye, the concentration of ions should apparently exceed a certain limit. Schenck enunciates the hypothesis that emanations of radioactive substances are nothing else than ozone. An attempt was made to account for excited radio-activity by the action of ozone.

Winkler<sup>1</sup> took a rather radical position, insisting that all of the reported radio-active elements simply contain a variable amount of radium, and furthermore he intimated that radium itself is not an element but that it may be impure strontium with an excessive electrical charge.

Davis,<sup>2</sup> in endeavoring to decide between the two hypotheses to explain radio-activity—namely, "Atomic Degradation" and "Molecular Change" (Armstrong and Lowry, Proc. Roy. Soc., 1903)—found that metallic selenium affected the photographic plate through black paper. Similar results have been reported by Taudin and Chabot.<sup>3</sup>

McCoy<sup>4</sup> discussed the decomposition of radium from the standpoint of the law of mass. The order of decomposition was considered as follows:



The radio-activity of an ore would be in proportion to all of these, but may be judged by the amount of uranium present, as was pointed out by Boltwood.<sup>5</sup>

Before giving the theories of those who have done most, experimentally, (Rutherford and his co-workers), toward an

1. Berichte 37, 1655 (1904).

2. Nature 70, 506 (1904).

3. Phys. Zeit., Aug. 25 (1904).

4. Berichte 37, 2641 (1904).

5. Am. J. Sci. 18, 97 (1904).

elucidation of the difficult problem, it is appropriate to mention several interesting facts bearing upon the subject.

In 1895, Perrin,<sup>1</sup> as did J. J. Thomson,<sup>2</sup> two years later, offered proof that the Crookes's rays consist of negatively electrified material particles. Omitting the mathematical calculations, it may be said that these particles carrying a unit charge are one ten-thousandth of a milligram<sup>3</sup> in mass. Thomson proved that the electric charge on a particle in case of gaseous electrolysis is the same as that of a hydrogen atom in liquid electrolysis; namely,  $1.13 \times 10^{-20}$ , E. M. U. and since the mass of hydrogen required to carry a unit charge, in the case of ordinary electrolysis, is one-tenth of a milligram, it follows that the mass of cathode particles required to carry a unit charge can be only one-thousandth as great. Therefore, the masses of these electro-negative particles constituting the cathode rays are one-thousandth of a mass of a hydrogen atom. The accepted value of the mass of a hydrogen atom is  $2.3 \times 10^{-21}$ , the mass of the cathode particles is  $2.3 \times 10^{-24}$ , milligrams. Thomson also determined the speed of the particles as being from 2.2 to  $3.6 \times 10^9$  c.m. per second. The speed of light is  $3 \times 10^{10}$  c.m. per second. Hence the cathode rays have a velocity of one-tenth of that of light. These velocities vary somewhat in different experiments, the highest value obtained being about two-fifths the speed of light. Thomson called the particles "corpuscles," as his ideas resembled Newton's corpuscular theory. In fact, Thomson suggests that they constitute negative electricity, which is a return to the single electric fluid theory of Franklin.<sup>4</sup>

The energy of the corpuscles is enormous. Although they are minute, the energy effect is considerable. \*It has been estimated that the energy emitted from each square centimeter

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1. Compt. Rend. 121, 1130.

2. Phil. Mag. V, 44, 293.

3. Phil. Mag. V, 48, 547 (1898).

4. Harper's Magazine, 103, 64, Sept. (1901).

of radium would melt a layer of ice of the same area and one-quarter of a mile thick in a million years. Or, as Lord Kelvin has said, the emission of matter and corresponding loss of energy have apparently been going on indefinitely in the past. As he says, it appears to place the first question mark after the great fundamental law of the conservation of energy.

Strutt devised an instrument which gives the nearest approximation to perpetual motion so far observed. The description of a radium clock as constructed by Mr. Harrison Martindale of England is given as illustrative of the principle of Strutt's apparatus. The registration of time is made in two-minute beats. The function of the apparatus is to exhibit the dissipation of negatively charged  $\alpha$ - and  $\beta$ -rays by radium. A small tube containing a minute quantity of radium is supported in an exhausted glass vessel by a quartz rod. To the lower end of the tube is attached an electroscope formed by two strips of silver foil. The leaves diverge, striking the walls

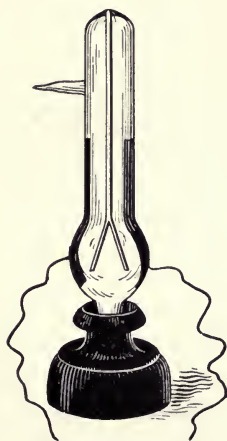


Fig. 46.

Strutt's Radium Clock. It has been suggested that a very reliable time-piece may be constructed on this principle. So far however no satisfactory method of mechanically registering the charging and discharging, that is, beats, has been devised.



of the vessel, which are grounded by wires, and are discharged. The operation is repeated incessantly requiring two minutes. In this instance it is calculated that it will take 30,000 years until the radium is exhausted.

The speed with which the corpuscles move from the radio-active substances is even greater than that of Crookes's rays. Becquerel has shown that their speed may be as high as two-thirds that of light. Other investigators have obtained even higher velocities,  $2.8 \times 10^{10}$  c.m., having been measured. The rays emitted by radio-active substances consist in part at least of material particles having a high velocity. Therefore, a loss of matter must go on continuously. Madame Curie estimates that radium emits from each square centimeter of surface 1.2 milligrams of matter in one million years. Therefore, it would be impossible to observe, by the most delicate balances at present available, any loss in mass during two million years.

Two important questions present themselves to the reader if these statements even approximate the truth. Where does this radium and radio-activity come from? and what is its real influence in the world?

W. E. Wilson,<sup>1</sup> Darwin<sup>2</sup> and Joly<sup>3</sup> independently suggested that radium might enter as an important factor in contributing to solar radiation and the maintenance of solar temperature. Reference has already been made to Rutherford's belief that the amount of radium present and uniformly distributed throughout the earth would be sufficient to account for its loss of heat. Thus it will be seen that the life of the earth has continued sufficiently long to allow the time necessary for the processes of evolution of the geologists and biologists.

Thomson, at a recent meeting of the British Association, as a result of his experiments on the universal distribution of radio-activity, concluded that each metal gives out a specific radiation which differs in its properties from the radiations sent

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1. Nature, July 9 (1903).

2. Nature, Sept. 24 (1903).

3. Nature, Oct. 1 (1903).



out by any other substance, and appears not to be a secondary radiation due to contact with some other form of radiation present in the atmosphere.

So far no satisfactory experimental evidence has been offered to prove that the energy of radium is derived from external sources. Yet the following idea gave the writer momentary comfort. An insulated wire formed into a circle, the ends free, horizontal, vertical, or inclined as far as the earth's surface is concerned, is perfectly neutral. Let it be revolved and an electric current is produced. Evidences of energy are had. Vary the speed and number of circles and differences in the current are observed.

Is it too great a draught upon the imagination to think that the atoms which are heaviest possess this same power? Their motion, perhaps on account of their weight, is such as converts the earth's lines of force into perceptible energy. The Curies early made a suggestion somewhat similar to this; namely, that radium might have the power of absorbing a species of Röntgen rays from the earth and converting them into other forms of energy. It has been shown that there is in fact a kind of penetrating rays, like the  $\gamma$ -rays of radium, near the earth's surface. The writer's dream was dissipated by finding a heavy constituent, inactive thorium. It comes back now, since Rutherford has shown that during the disintegration of the emanation a temporary inactive state is arrived at.

So substances may vary in the amount of their activity, as shown for radium and thorium preparations. Fresh radium requires time to reach its maximum. So does thorium, while Rutherford's thorium-X runs down to a minimum, which is the maximum of its mother substance. In one case it requires time to reach the stable speed. In the other, it requires time to slow down to the safe equilibrium rate. The dynamo analogy becomes more perfect when we have active and inactive thorium.

J. A. Alexander<sup>1</sup> insists that radio-activity is due to external energy. He says:

"All matter, as we know, is continually *receiving* and *giving out* energy but the total sum of the plus and the minus in the universe equals zero.

Radio-activity and magnetism are in some respects analogous. Each is exhibited most strongly by *one* element, and to a lesser degree by several closely allied elements. Each can be communicated to some other bodies without apparent loss to the original active substance. Both are impaired by heat, fusion or solution, which seem to alter the conditions of the molecular complexes. We believe magnetism to be consequent upon the localization of ever-existent cosmic forces; and it seems to be probable that radio-activity can be traced to the same origin."

Rutherford and Soddy, assuming that radio-activity is an atomic and not a molecular property, advance the most acceptable theory yet put forward; namely, the atoms of radio-active elements undergo spontaneous disintegration. This takes place in fixed and well-marked steps. These changes are nearly always accompanied by the emission of  $\alpha$ -rays.

The emission of the radiations is dependent solely upon the amount of active element present. The rate of emission is not affected by variations in temperature or by any known chemical or physical forces. It has been demonstrated that the radiations consist for the most part of positively and negatively charged particles, projected with great velocity. Hence it has been assumed that part of the atoms escape from the atomic system. It is difficult to imagine that the projected particles can suddenly acquire such a velocity of movement through the action of force, either within or without the atom. To illustrate this point, attention may be called to the fact that the  $\alpha$  particles, according to Rutherford, "would have to travel from rest between two points, differing in potential 5.2 million volts, in order to acquire the kinetic energy with which

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1. American Chem. Society (N. Y. Sect.), Nov. 11, 1904.

it escapes. They must, therefore, escape from a system which is already in exceedingly rapid motion. Consequently, the energy exists before hand in the atoms from which they escape."

J. J. Thomson, Larmor and Lorentz have urged the conception that the atom is very complicated, being made up of charged particles, in rapid oscillatory or orbital motion. As the particle is atomic in size, it must be composed of electrons in motion. The radio-active elements, therefore, are composed of positively charged particles, whose mass is about that of hydrogen or helium.

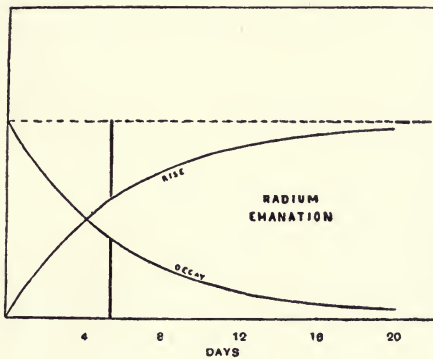


Fig. 47.—CURVES SHOWING DECAY OF ACTIVITY OF THE EMANATION AND RECOVERY OF ACTIVITY OF RADIUM (AFTER RUTHERFORD).

The curves (see Fig. 47), showing the decay of the activity of the emanation and the recovery of the activity of radium are extremely interesting. It will be noted that they are complementary to each other. When the emanation has lost one-half of its activity, the radium has spontaneously regained one-half of its lost activity. The sum of the three factors, namely the activity from the separated emanation, the activity of the remaining radium, and that lost, constitute a constant. This



is accounted for by assuming that radium is always manufacturing fresh emanations at a definite rate. When the emanation is removed, the radium is temporarily exhausted but immediately proceeds to produce more emanation and store it up. As these two reach an equilibrium, we have the constant activity of the radium.

The laws which control the material  $\alpha$  and  $\beta$  particles are different from those of ordinary chemical changes. Temperature, which plays an important part with all ordinary chemical reactions, has no noticeable effect in changing the processes occurring in radium, as already referred to. The rate of decay of the activity of the emanation is apparently not changed by severe physical and chemical treatment. Assuming that changes occur within the atom, we should expect temperature to have little influence, for we know from our experience with different elements that wide variations in temperature have little effect in altering stability. During this process of disintegration at least five distinct substances are produced. The emanation is, chemically, an inert gas, while the other products act like metallic substances—soluble in some acids and volatilized by heat. Each of these different substances is different from an ordinary chemical element, because it is not permanent and is continuously and rapidly changed into another kind of material. This is shown graphically by Rutherford in the accompanying diagram (Fig 48). Rutherford gives a very interesting table showing the time required for the different changes.

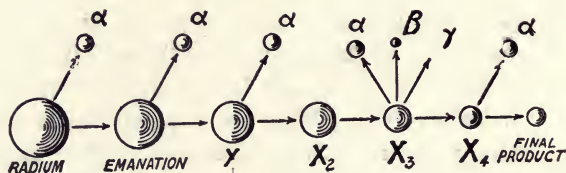


Fig. 48.—DIAGRAM TO REPRESENT THE DISINTEGRATION OF A RADIUM ATOM (AFTER RUTHERFORD).

Name of Substance	Time	Remarks
Radium		
v		
Emanation	4 days	1st product
v		
Emanation X (1st change)	3 minutes	2nd product
v		
Emanation X (2nd change)	21 minutes	3rd product
v		
Emanation X (3rd change)	28 minutes	4th product
v		
Emanation X (4th change)	very slow	5th product
v		

In each case, but one, the transformation is accompanied by the throwing out of  $\alpha$  particles and in one only, namely the fourth stage, are evidences of  $\beta$ - and  $\gamma$ -rays obtained. Some evidence is already had which indicates that radio-tellurium is really the fifth product of the disintegration of the radium atom.

Each one of these chemical products has distinct chemical properties, which distinguish it, not only from its immediate neighbors but from the parent element and the final product.

It has been calculated that the weight of the emanation obtained within four days from one gram of radium bromide is about 1/100 of a milligram, while the weight of the fourth product, which breaks up in twenty-eight minutes, is about 3/100,000 of a milligram. This amount is entirely too small to be detected by balances, so it can scarcely be hoped that enough of it can ever be collected, in sufficient quantity, on account of its limited life. The inactive products, however, will continue to increase as long as there is any of the mother element present. This is really an apparent case of the transmutation of the elements.

Truly, as Runge says, "Nature is becoming more and more disorderly every day."

In the author's humble opinion we are not yet warranted in accepting this as the correct solution of the problem, beautiful as the explanation is. So far, however, nothing better has been offered and, as with all things in science, it should be accepted until something better takes its place.



Ramsay<sup>1</sup> states that electrons are not matter but are capable of causing profound changes in matter. For a year a solution of radium bromide was kept in three glass bulbs, each bulb connected to a Töpler pump by means of capillary tubing. This was done to collect *ex-radio*, the term he proposed for "emanation-substance." Each of the bulbs, to avoid accident, was surrounded by a small beaker, one consisting of potash glass and the other two of soda. The potash beaker became brown, while the two soda beakers became purple. This variation in the color was attributed to the probable liberation of the metals potassium and sodium, which ordinarily exist in that very viscous liquid, glass, in the colorless ionic state. The glass had not been subjected to the  $\alpha$ -rays, therefore, to no bombardment of what is usually called matter except the molecules of the surrounding air. The colored beakers are radio-active and the radio-active film dissolves in water. After careful washing the glass was no longer radio-active. The solution contained an emanation, for in bubbling air through it and cooling the issuing gas to  $-180^{\circ}$  C, part of the radio-active matter was retained in the cooled tube. This air, also, discharged an electroscope. The period of decay was very rapid. In having such a short period of existence the emanation resembles that of actinium. The water solution on evaporation gave a residue which was strongly active. On adding mercurous nitrate to the dissolved residue and then adding hydrochloric acid the greater part of the active matter was thrown down with the mercurous chloride. This appears to indicate the formation of an insoluble chloride. The activity of the mercurous chloride remained unchanged for ten days. The filtrate from the mercurous chloride was active. On precipitation the mercurous sulphide was also active but its activity decayed in one day. The filtrate from that gave an inactive precipitate with ammonium hydroxide, hence the active matter forms an insoluble chloride and sulphide. These, when dissolved in aqua

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1. "Present Problems of Inorganic Chemistry," address before the International Congress of Arts and Science, St. Louis, 1904.

regia, gave an insoluble sulphate when barium chloride and sulphuric acid were added. This indicates the formation of an insoluble sulphate, that is a body somewhat resembling lead. The explanation given for this was perhaps misinterpreted by the secular press into the actual building up of elements; in short, a verification of the dream of the Alchemists, although Ramsay gave as his "guess" that such an explanation was more than likely.

Without doubt the most valuable of the recent work on the "Transformation Products of Radium" was that reported by Rutherford<sup>1</sup> at the International Congress at St. Louis. He studied the residual activity of a bismuth rod exposed to the emanations of radium. The residual activity consists of both  $\alpha$ - and  $\beta$ -rays, the latter being present in unusually large proportion. He, also, noted the proportion of the  $\alpha$ - to  $\beta$ -rays from a platinum plate one month after removal from exposure to the emanations. Unlike the  $\alpha$ -rays activity, the activity measured by the  $\beta$ -rays remains constant, consequently the proportion of the  $\alpha$ - to the  $\beta$ -rays steadily increases. The intensity of the  $\beta$ -rays did not vary much over a period of nine months. This want of proportionality between the  $\alpha$ - and  $\beta$ -rays shows that the two types arise from different products. The activity deposited apparently consists of two kinds of matter: (1) a product giving only  $\beta$ -rays which is soluble in sulphuric acid but not volatile at  $1000^{\circ}$  and which is not deposited on bismuth; and (2) a product giving out only  $\alpha$ -rays which is soluble in sulphuric acid, volatile at  $1000^{\circ}$ , and is deposited from a solution on bismuth.

The  $\alpha$ -ray activity increases if the  $\beta$ -ray product is present. It remains sensibly constant, or generally very slow in decay, if the  $\alpha$ -ray product is removed from the  $\beta$ -ray product by the action of the bismuth plate. The  $\beta$ -ray activity remains sensibly constant independent of the presence of the  $\alpha$ -rays. These results show that the  $\beta$ -ray product is the parent of the  $\alpha$ -ray product. The amount of residual activity from radium

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1. Phil. Mag. 8, 636 (1904).

	MASS	VELOCITY	ENERGY
$\alpha$	○	—	⊗
$\beta$	•	————	•

Fig. 49.—A graphic comparison of the  $\alpha$  and  $\beta$  particles. The velocity is represented by the length of a line and the mass and energy by spheres.

emanations depends upon the amount of the emanation present and the time of exposure to the emanation. Rutherford has changed his nomenclature and illustrates graphically the change, as shown in Fig. 50. By such an explanation he is able to account for the presence of radium-D and radium-E in pitchblende. He doubts if radium-D has been separated from pitchblende, although it is barely possible that the radiolead of Hofmann, which emits a large amount of  $\beta$ -rays, may be radium-D. Concerning radium-E, he thinks there is little doubt that it is the radio-tellurium of Marckwald, as his active bismuth gave out only  $\alpha$ -rays. Rutherford states that it will be of extreme scientific value if the radium-D can be had from pitchblende, as it could be used for many of the purposes of radium. Its activity is about 25 times that of radium and the rate of change in the activity is sufficiently slow to be negligible for most experiments.

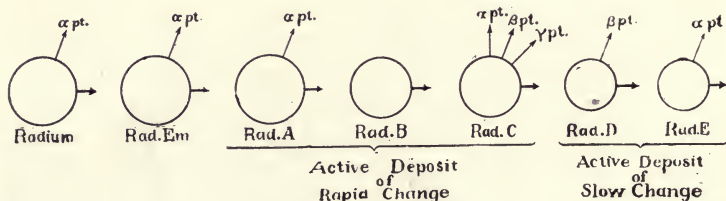


Fig. 50

Diagrammatic representation of the changes occurring in radium and its emanations according to Rutherford [Phil. Mag. 8, 641 (1904).]

## CHAPTER VI.

THE PHYSIOLOGICAL ACTION OF RADIO-ACTIVE  
SUBSTANCES AND THEIR THERAPEUTIC  
APPLICATIONS.

From what we have learned in the preceding chapter as to the resemblances among the radium and other rays, it is not unreasonable to anticipate specific physiological effects from the radio-active substances. That this is true, however, was accidentally and painfully discovered previous to the observation of Walkhoff<sup>1</sup> that radium rays inflame the skin similar to the Röntgen rays.

Becquerel carried a small tube of an impure radium preparation in his vest pocket for six hours. A few days later he observed a reddening of the epidermis of the abdomen opposite the location of the pocket in which he had placed the radium compound. It was not long before the inflammation became pronounced, and an ulcer developed which required several months for the healing.

Giesel<sup>2</sup> exposed the inner portion of his arm, for two hours, to 0.27 gram of a radium preparation, enclosed with a double celluloid capsule. After two or three weeks the skin reddened, blisters formed and the epidermis peeled just as with a burn. The growth of hair was also destroyed and did not come out anew, although a smooth white skin reformed. These observations were verified by Becquerel and Curie. "The action of radium upon the skin can take place across metal screens, but with weakened effect."<sup>3</sup> (Fig. 51.)

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1. Photogr. Rundschau, Oct., 1900.

2. Ber. d. deutsch chem. Ges. 33, 3570 (1901).

3. Madame Curie's Thesis.



Fig. 51.

Professor Curie's arm, showing a scar resulting from a radium sore. (Through the courtesy of the Success Company.)

Rehns studied the precise effect of radium burning upon the skin. The rays from twenty milligrams produced no pain and left no mark at the time of the application. A red mark appeared 24 hours later, remaining for two weeks and then fading away, leaving a scar similar to a burn. If the application be continued for ten minutes the mark becomes visible in 18 hours, but ulceration does not occur unless the radium has been applied for at least an hour. If the burned spot be treated medicinally the wound may be cured in six weeks, but if not attended to, it ulcerates, becomes painful and the ulceration lasts for an indefinite period.

Moles can be destroyed by the application of radium for ten minutes.

Abbe<sup>1</sup> appears to have been the first to record the fact that an ordinary wart (*verruca vulgaris*) is caused to disappear by the application of radium. The age of the growth seems to have no influence. Within three or four days a pink zone appears around the base of the growth, then it begins to flatten and usually disappears inside of ten days, leaving a smooth skin.

Giesel observed the action of radium upon plant growth, noting that the leaves treated turned yellow and withered away. He, also, discovered the action of radium upon the eye. If a radio-active substance be placed near the eye or

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1. Medical Record, 66, 321 (1904).



temple, when the person is in the dark, a sensation of light is experienced. On the announcement of these observations the secular press hailed a cure for blindness. Heinstadt and Nagel and Crzellitzer, however, have studied the phenomena carefully and demonstrated that the centre of the eye is rendered fluorescent by the action of the radium. This gives the sensation of light experienced.

The effect upon the eyes produced by radium is a diffuse brightness, somewhat like that one experiences when he steps from a dark to a brilliantly lighted room, with the eyes slightly closed, that is, the interior of the eye begins to fluoresce. The cornea, the lens, especially the vitreous humor, and perhaps the retina are involved. This is quite different from the effect of Röntgen rays, which act upon the retina alone. A pure radium salt acts with such intensity that the effect may be obtained by placing the chemical back of the head, and without the intervention of the optical apparatus at all. The Becquerel rays may produce an apparition, but it is not possible to secure a picture as they are deficient in a characteristic property of visible light, namely, refraction.

Concerning the statement that totally blind persons are not only able to see the radium light, but perceive the phosphorescent radium screen and distinguish silhouettes, coins, keys, etc., placed on the screen, Halzknecht and Schwarz<sup>2</sup> offered two explanations. The radium rays passing through the tissues reach and irritate the optic nerve and stimulate the relics of visual capacity left in it. In this case the nerve would also experience the same tendency. The blind person, after a little practice, should be able to perceive any dark object on a bright background. Heller, having made some experiments along this line, found it possible and that the same results could be obtained by any light. The radium had

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1. See Karewski in Marckwald's "Über Becquerelstrahlen und radio-active Substanzen," *Moderne Arztliche Bibliothek*, Heft 7, Berlin (1904).

2. "Ueber Radium-strahlen," *Wien Klin. Wochenschrift* 16, 25.

nothing specific to do with the phenomenon. The other and perhaps the correct explanation, is based upon the transformation of the energy of the radium rays into objective phosphorescence. Animal tissue, hair, bone, muscle, drops of water, etc., are rendered more or less vividly phosphorescent under the action of the radium rays. The radium rays render the sclerotic phosphorescent and this phosphorescence is seen by the point of the retina opposite to it. When a visual sensation is experienced from compression of the eye-ball, the source of the light is referred to the point opposite to that from which the compression light proceeds. This is the reverse of the experience with radium.

There is no doubt but that blind people whose retinas are intact are sensitive to the action of radium, but those with diseased retina experience absolutely no luminous sensation. London, in St. Petersburg, aroused many hopes by his observations, but Greeff, in Berlin, on extending the experiments, came to the same conclusions given above.

Javal<sup>1</sup> has suggested that blindness with alteration of the retina can be distinguished from that due to glaucoma or corneal opacity, because patients with the latter condition see radium rays as well as do those of sound vision.

Rollins<sup>2</sup> suggested the use of radio-active substances as a substitute for the X-light. He prepared a capsule, with an aluminum front and back of comparatively non-radiable metal which could be worn over a lupus or superficial cancer.

Danlos<sup>3</sup> of the St. Louis Hospital, Paris, apparently was the first to apply radium in the treatment of certain affections of the skin similar to the treatment with Röntgen and the ultra-violet rays (Finsen). A case of lupus of the face was

1. *Revue Internationale d'Electrotherapie et de Radiotherapie*, Nov. and Dec., 1902.

2. *Medical News*, Jan. 25 (1902).

3. *Revue l'Electrotherapie et Radiotherapie*, Nov. and Dec. (1902).  
*Ann de Dermatologie et de Syphilis*, July (1902).

treated with radium chloride (19000X). The disease disappeared with the formation of a smooth white cicatrix, blending into the surrounding tissue.



Fig. 52.

Dr. Danlos and assistants treating a lupus patient with radium. (By courtesy of the Success Company.)

Hallopeau and Gadaud<sup>1</sup> report that too prolonged application of radium led to atonic ulceration which lasted for five or six months; also, that ulcers of normal tissue can be avoided by proper technique and care.

Blandamaur has also used radium in lupus.

Danycz<sup>2</sup> found that radium destroys the skin of guinea pigs and rabbits; but subcutaneous and muscular tissue do not seem so sensitive as skin. Nervous tissue is sensitive to its action. A glass tube containing a radium salt, which was placed against the skin over the spine, produced death in young animals. In older animals, the osseous tissue seems to protect the cord

1. Ibid.

2. Compt. Rend. 136, 461 (1903).

against the radiations. Dancyz and Böhm showed that various larvae and embryos are profoundly modified in their growth, many being killed, when subjected to the radiations; others developing into monstrosities, because of unequal stimulation. The latter observer found that the radiations exercise an especially intense action on tissues and cells in proliferation. Non-fertilized eggs may undergo more or less parthenogenetic development and give rise to atypical formation. In the case of some animals, where the skin has been burned by the rays, the hair appears to be forced into rapid growth. It seems that various effects are obtainable, depending on the tissue or cell exposed, as well as on the quantity and quality of the rays.

When two groups of meal worms were placed in jars, over one of which radium was suspended, many of the radiumized worms died; those which lived showed much retardation. The worms in the parallel jar passed through the regular cycle of life, laid eggs which grew to worms, and repeated the cycle three or four generations. The radiumized worms still remained mere worms.

Böhm<sup>2</sup> reported, as a result of experiments, that lower organisms are quickly destroyed by radium rays.

Tur exposed eggs, for 24 to 70 hours, to the action of a 35 per cent. radium chloride. The central parts were particularly affected, the surrounding blastoderms remaining untouched. Aside from numerous variations in the embryonic skeleton, there was a peculiar vascular formation in the centre of the embryo and other phenomena, showing a peculiar localization of the injurious radio-active effects.

Holzknicht<sup>3</sup> reported psoriasis and lupus hypertrophicus as cured by both X-rays and radium. The radium was superior, if anything. Epithelioma of the cheek also rapidly subsided. Apparently the healthy skin in the neighborhood of

1. Compt. Rend. 136, 1016 and 1085.

2. Soc Biol 55, 1655.

3. Wirkung der Radiumstrahlung bei Hautkruheiten, Vienna Klin Wochenschrift, 16, 27 (1903).



these affections is not seriously interfered with. Radium seems to produce degenerative processes in the cells of the intima of the blood vessels, shown by Scholtz, as characteristic of the Röntgen rays. On account of the degeneration, there ensues a rapid dilatation of the capillary and precapillary vessels. These observations were made upon a remarkable case of telangiectesis.

A committee appointed by the Vienna Academy of Science to investigate the results of the treatment of cancer with radium, reported, says the *Popular Science Monthly*, "in nine cases in which the treatment was used abatement in the cancerous swelling resulted, and in two of these cases the swelling had not reappeared after five months' time. A case of cancer of the palate was much improved by the treatment. The use of radium is not recommended when an operation is practicable." Numerous other cases of the beneficial results of the radium treatment have been reported. The press reports from the London Cancer Hospital do not appear to be so encouraging.

Exner<sup>1</sup> applied a capsule containing a radium preparation by fastening it to the spot with adhesive plaster. The nodules following an operated melano-sarcoma disappeared when treated twenty-five minutes with the radium. They disappeared before the superficial tissues exhibited necrosis from the action of the rays. A capsule containing radium bromide, protected from moisture by a rubber cot, was applied to a case of epithelioma, at the corner of the mouth, six times within seventeen days. The tumor perceptibly diminished, the ulcer began to heal over; at the end of the month it had apparently vanished. This physician also reports on the radium treatment of six cases of carcinoma of the oesophagus.<sup>2</sup>

The technique was the introduction of a scrap of radium embedded in dammar, and fastened to a No. 16 sound. The

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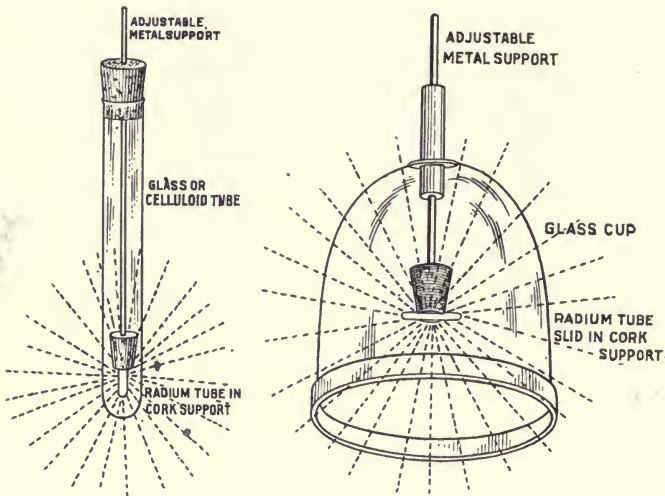
1. Radium Treatment of Malignant Tumors and Cutaneous Affections; Vienna Klin Wochenschrift 16, 27 (1903).

2. Semaine Medicale. Paris, 24, 9 (1904).



increased permeability of the structure noted in the five cases was probably due to necrosis of the structure tissue under the influence of the radium, thus giving permanent results.

Morton<sup>1</sup> favors radiation to operation in the treatment of malignant disease in its earlier stages. Robert Abbe<sup>2</sup> summarized his wide experience with radium in the treatment of



Figs. 53 and 54.

Method of applying radium preparations in focal treatment according to Morton.

lupus, epithelioma, rodent ulcer and carcinoma, by saying "lupus can usually be cured by a few applications of radium, varying in number, and frequently with the strength of the specimen. Superficial epithelioma, rodent ulcer, and small recurrent cancer nodules, can be caused to disappear by cautious application, but if mild preparations are used, very little effect is seen. Indeed, the judicious use of Röntgen rays is more efficient along the same line in results and with only brief applications."

1. "Treatment of Cancer by the X-rays with Remarks on the Use of Radium;" International Journ. of Surgery, New York, Oct. (1903).

2. Yale Medical Journal, June (1904).

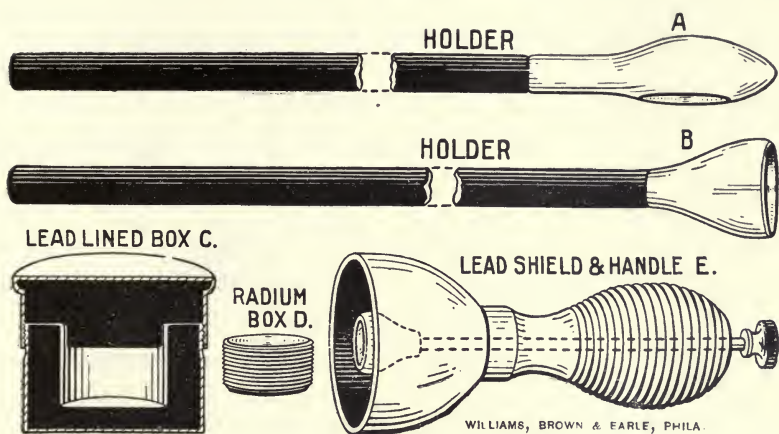


Fig. 55.

Apparatus of Williams, Brown & Earle, used in applying radium compounds in medicine.

Seventy-five milligrams of radium, in a mica-covered box, were bound to Goldberg's<sup>1</sup> arm for three hours. Four days later a red patch developed, changed into a necrotic ulcer on the fourteenth day, and other ulcers developed on different parts of the arm; also, on the skin, in the groin and hand. The healing processes commenced first in the later patches. The ulcers were slow but sure in the healing. The action of the radium was probably due to its activity and not to its bulk. The exposure and the subsequent phenomena were painless. The necrosis developed without fever, and the ulcer had a peculiar morbid character. Rodent ulcers were cured.

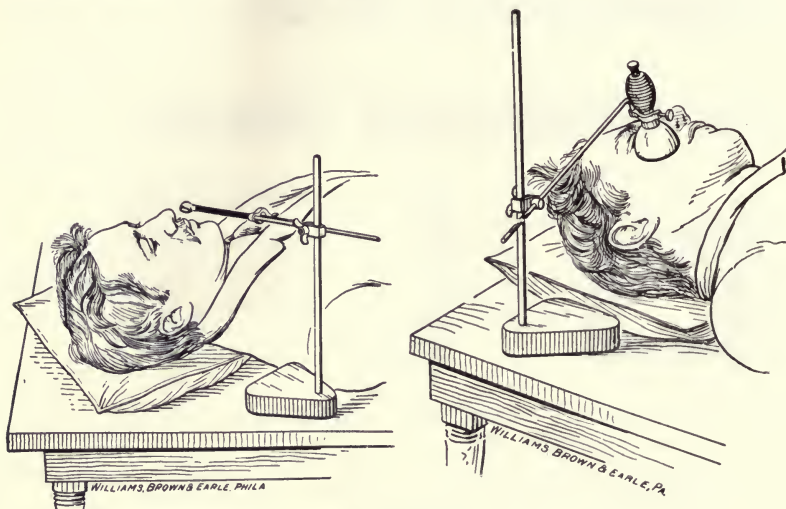
Cleaves<sup>2</sup> reports the cure of several cases of recurrent epithelioma of the rodent ulcer type.

Williams, who has carried out most systematic investigations on the medicinal applications of radium compounds, calls

1. "Zur Frage der Beziehungen zwischen Becquerelstrahlen, und Hautaffektionen," Goldberg and London, *Dermatologische Zeitschrift*, Berlin, 10, 5 (1904).

2. 13th Ann. Meet. Amer. Electro-Therap. Assoc., Atlantic City, Sept. 24, 1903.

attention to the fact that radium possesses less value in diagnosis than X-rays, as it does not differentiate so clearly. He



Figs 56 and 57.

The simple technique of applying radium compounds in the treatment of skin diseases.

employed it as a therapeutic agent in nine cases of skin affections, two of eczema, and psoriasis, four of lupus vulgaris, and one of acne. Success was not so good in eczema, although there was some improvement. In lupus, the results were satisfactory; also with acne. This important paper has reference to other work, which treats of thirty-three cases which show that radium is useful in treating some skin diseases and superficial new growths, including in this class those of the cervix uteri. He endeavored to differentiate as to the value of the different rays by isolating the  $\beta$  and  $\gamma$  rays. The burning power he attributes to the  $\beta$  rather than the  $\gamma$  rays. As a result of his large experience, he regards the therapeutic action of radium as being of greater value than the X-rays, excepting that the latter is able to cover larger areas. He concludes as follows: "If the results obtained by radium prove

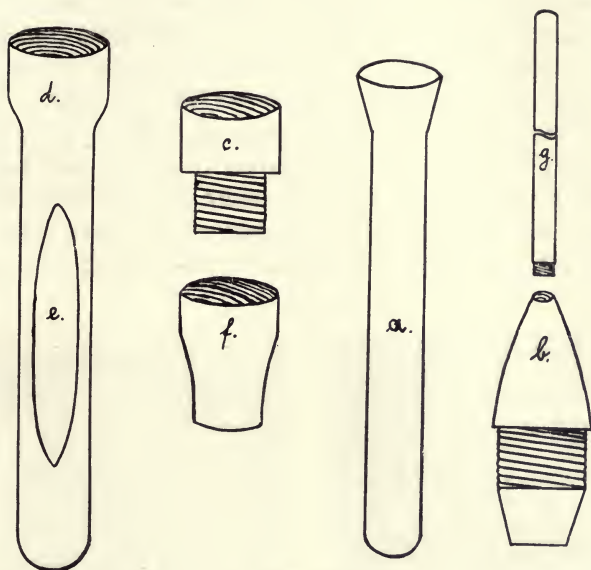


Fig. 58

Lieber's Aluminum Tube for containing Radium.

A.—Aluminum tube containing radium, which is closed hermetically by

B.—a wedge fitted with a screw thread so that

C.—a lid may be screwed on same, thereby closing the tube hermetically. This must not be opened after the radium has been filled in.

The lid C has a screw thread on which may be fastened  
D.—a silver mantle or cover, which can be removed at will, or in which holes or windows of any desirable size may be cut, such as indicated in E, to permit the escape of all radiations.

F.—is a short silver mantle which is to be used to produce a smooth, ending surface by attaching same to C when the long silver mantle D is not to be used.

G.—is one of the great variety of handles which may be readily attached to B.

There is also furnished a small plug, which has on its lower end a screw thread, which will fit readily in B. To this plug may be attached thin rubber hose: Catheters, Bougies, etc., to answer any purpose.

permanent this new therapeutic agent will be largely used instead of the X-rays; but the two will supplement each other. Certain diseases promise to yield more readily to treatment by radium, and other diseases more readily to X-rays. A disease that has attacked different parts of the body of a given patient may be better treated in certain regions by radium, and in others by the X-rays, and it is quite possible that in some cases the two remedies used together on the same area, and at the same sitting may accomplish better results than either alone."<sup>1</sup>

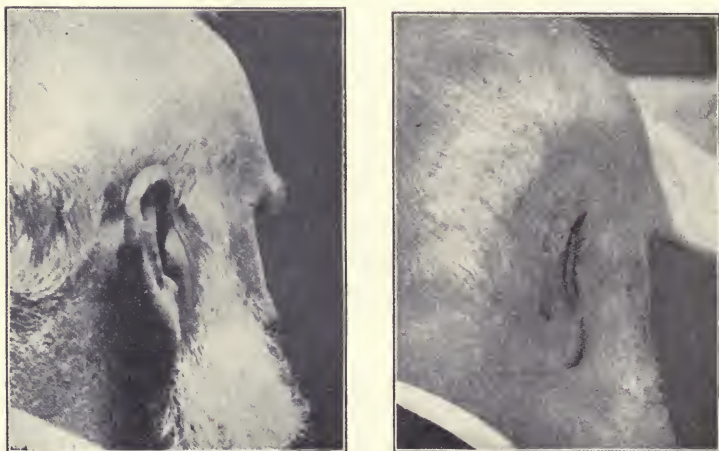


Fig. 59

This shows an epithelial cancer of the ear, before and after treatment by radium. The disease remained cured after one year. (Robert Abbe in *Medical Record*, 66, 321, 1904.)

Truman Abbe<sup>2</sup> remarks, "The radium rays, no doubt, should be classed with the X-rays, the Coley serum, Adamkiewicz serum, and the various caustics. These have given cures

1. "Some Physical Properties and Medical Uses of Radium Salts; with a Report of Forty-two Cases Treated by Pure Radium Bromide," F. H. Williams, *Med. News*, New York, Feb. 6 (1904).

2. *Washington Med. Ann.* 2, 363 (1904).



in a few cases of inoperable and malignant diseases, but they are far too uncertain to be used except when operation is out of the question."

Lyster, of the Middlesex Hospital, uses radium of low activity and pitchblende. He applies pitchblende directly to the diseased structure for twenty-four hours, binding it on. The radium is permitted to excite only the granulations.

At the Cancer Hospital, London, radium is used in an apparatus made of ebonite with a quartz shield. To condense the radiations the shield is held against the ulcer.

MacIntyre<sup>1</sup> applied radium by enclosing it in a small cell with a mica face. This was surrounded with a small piece of India rubber tube which fitted into the apex of a glass cone. This localized the action of the radium to the particular part to which it was applied.

According to Robarts<sup>2</sup> the treatment is done through a rubber pocket. He remarks: "It has been observed that higher activity gives better results than lower activity."

David MacKenzie<sup>3</sup> does not claim for radium any special value over the X-rays. Phimosis scytitis is increased and ruga scytitis varies, being increased within the neighboring walls of the vessels. He also reports the curing of rodent ulcer, tuberculosis, verucca, cutitis, rodent cancer and the disintegration of moles. Fragmentation of the covering of coloring matter was observed, as after X-ray treatment. The effect of radium is more rapid than that of the Röntgen rays; that is, a tissue reaction is quicker. He, also, states the method of application as used; that radium was not used in carcinoma; and suggests the possibility of applying thorium in large quantities to septic ulcers.

Sichel<sup>4</sup> applied five milligrams of radium bromide forty-two times to rodent ulcer with success.

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1. British Medical Journal, June 6, July 25 (1903).
  2. American Journal Surgery and Gynecology.
  3. British Medical Journal, Jan. 22 (1904).
  4. British Medical Journal, Jan. 23 (1904).

Apolant<sup>1</sup> studied the retrogression of carcinoma on mice under the influence of radium rays. The carcinoma cells vanished and there was proliferation of the connective tissue. In addition to the destructive action, it appeared to induce specific absorption of dying carcinoma cells. He remarked that the loss of penetrative power imposed such a limit to the effect of the radium treatment, that it has rendered it dubious whether as a therapeutic agent, it has much of a future.

Pozzi and Zimmern<sup>2</sup> reported an improvement in the case of cancer by treatment with radium. They, also, called attention to the necessity of determining the extent of the dosage.

Darier<sup>3</sup> noted the rapid and penetrative analgesic action of radium in certain cases of cyclitis and irido cyclitis. Preparations of low intensity were often capable of rapidly removing pain. In two cases of convulsive neuralgia, which came on frequently, the attacks ceased after a few applications of radium to the temple for two or three days. Radium effected a cure of an acute facial paralysis of recent origin.

Foveau de Courmelles<sup>4</sup> found by local application of radium chloride that the pain from facial neuralgia or cancer could be alleviated. He reports, also, that a plaster of thorium oxide may be successfully applied in the shape of a sort of varnish and the powder may be wrapped in tin foil and applied to the face. The successful treatment of several severe cases of neuralgia were reported.

Pusey<sup>5</sup> gives an excellent resumé of the therapeutic possibilities of radium. Concerning its effects upon the nervous system he says, "The symptoms are first depression of the central nervous system followed by *flexures* of the cerebro-spinal system. The explanation of these nervous symptoms lies in the

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1. Deutsche Medicinische Wochenschrift (1904).

2. Medecine Moderne, July 6 (1904).

3. Lancet, March 5 (1904); Paper presented before the French Academy of Medicine, Feb. 16 (1904).

4. Progres Med., May 28 (1904). See also his book on the subject.

5. Journ. A. Med. Assoc., July 16 (1904).

disintegration of the nerve cells produced by the Becquerel rays. The Becquerel rays affect at the same time the skin, epithelium, connective tissue and blood vessels. The effect on the last named appears first."

Holkin,<sup>1</sup> in a very thorough study of the action of the Becquerel rays upon the skin, noted the cellular degeneration and dilatation of the vessels in normal as well as in lupus tissue. The changes appeared only in the most superficial layers of skin.

From a comparison of Holkin's work and studies of Scholz on X-ray burns in young pigs, it is quite evident that the action of the two agree very closely, and may be said to be identical, with the single difference of the greater depth of action of the X-rays. The cells of neoplasm are as susceptible as new cells produced by irritation to the effects of the Becquerel rays, but they are of lower resistance, consequently their structure is disintegrated and they degenerate before the irritant cells are so violently affected.

Pusey insists that radium "will have a definite, though a limited field of usefulness in the treatment of regions situated in inaccessible locations, where it is difficult or impossible to apply the X-rays, but where radiations from radium can be applied readily." He reports its application on carcinoma of the uterus, rectum and mouth. He reports no definite effect from the use of thorium nitrate and oxide.

Schamberg<sup>2</sup> directs attention to the decided difference in the susceptibility of different individual radiations.

Bulkley<sup>3</sup> reported in one case of lupus better effects from the X-rays than with radium. He applied it successfully in case of epithelioma beneath the tongue and to the tonsil. Treatment by surgical means would have been difficult. The disease disappeared gradually under the influence of the radium.

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1. Archiv f. Dermatologie und Syphilis, 65 (1903).
  2. Journ. A. Med. Assoc., July 16 (1904).
  3. Journ. A. Med. Assoc., July 16 (1904), p. 180.

Plimmer<sup>1</sup> made a very searching investigation of seventeen cases of carefully diagnosed cancer in the Lister Institute of Medicine. Not for any case examined did he secure favorable results. "The radium had apparently no effect with regard either to cure or relief of pain." Several of the patients having died, careful microscopic examinations were made of sections of cancerous tissue. In all the cases examined no changes were found, either in the cancerous tissue or fibrous cell, and none were degenerated. According to him it appears as if the emanations from radium can only act upon young and growing cells, and the altered cells, especially if surrounded by old tissues, are less and less affected. If there is a succession of fibrous tissue, the cells are not at all affected. It is not clear that he used the most modern containers for the radium.

The etiology of cancer is not yet understood. It appears from the sifted evidence that thus far radium offers little hope as a permanent cure for the dreaded disease, especially after it has become deep-seated. It is generally accepted as a fact, however, that temporary relief from pain and a retardation of a cancerous growth may result from its application. Its portability, easy dosage, remarkably localized action, render radium a permanently valuable addition to the therapeutic arsenal, for the technique is simple.

The procedure of Williams,<sup>2</sup> for guidance of others, is here-with given: "When not in use the radio-active preparation should be kept in a thick lead box or envelope. When in use, preferably, all sides except that side next to the diseased tissue, should likewise be covered with thick lead to protect the operator. The compound should not be brought near photographic plates, unless it, or the plates, be within lead, as it would injure them."

"Method. The method of using the radiations from radium is simple. If the strongest action from the radium is desired, the metal box containing the salts is placed on the part to be treated; in this case the box should first be covered with a thin rubber cot, or other suitable substance, which can be readily

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1. The Lancet, Apr. 16 (1904).

2. The Medical News, N. Y., Feb. 6 (1904).



removed so that a new cot may be used for each patient and the old one burned up. By this means, the radium capsule does not come in direct contact with the part to be treated, but is separated from it by this new and clean covering. If a weaker action of the radium salts is indicated, the capsule should be placed at a greater or less distance, according to the needs of the case, the intensity of the rays diminishing as the square of the distance.

“Exposure. It is important to remember that an over exposure of a part may result in a burn, and that this burn may not become evident in several days after the exposure has been made. Further, that the exposures differ for different diseases, even superficial ones. Experience, therefore, is necessary to judge not only of the proper length of exposure, but also of its frequency.

“Length and Frequency of Exposures. Exposures must in some cases be longer, in others shorter, and the frequency with which they are given must vary. In some cases the treatment should be pushed; in others harm, rather than good, would result from this procedure. The exposure, then, must be adapted to the special case, and further experience is necessary to decide the best for all cases, but as a general rule, it may be said that when the beta and gamma rays of pure radium bromide (I have discarded the use of the weaker salts) are used together, for the treatment of superficial lesions, and the radium capsule is placed on the part to be treated, the length of the exposure should be ten minutes to one hour, according to what the practitioner desires to accomplish.

“Exposures should not be made every day. Two or three times a week seems to me the safer procedure, as by this method an interval is given during which progress can be watched.

“An exposure of many hours would be necessary if weaker forms of radium are used, that is radium of 1000 to 8000 activity, before any special results could be obtained, and these weaker forms would not be so efficient as compared with the pure radium. Pure radium bromide is none too strong for the work to be accomplished in certain cases; in those in which the full strength is not necessary, the radium capsule can be placed at any distance desired and the exposure can, also, be shortened.”

In X-ray treatment dermatologists are agreed that great care must be exercised as to idiosyncrasy of the patient, kind of tube, vacuum, strength of the current, length of application,



frequency, etc. (See Chapter VII.) It has been asserted that radium compounds of a definite strength may be used to obviate many of these unknown factors. Piffard<sup>1</sup> sounds a timely warning and most reasonably calls attention to the differences which exist between the actual radio-activity of *naked* radium and *efficient* radio-activity of a protected compound. "Radium affects the photographic film and also the electroscope and electrometer, but it is by no means certain that the radiations that are most active photographically are the ones that most strongly ionize the air in the electrical apparatus, and it is still less certain as to which is the most efficient in its action on the human tissues."



Fig. 60

Exact representations of a giant-cell sarcoma of the jaw of rapid growth during two months. On the left the sketch is before treatment. The teeth of the lad were so loose as to be readily removed by a string. On the right may be seen the improvement after six months' treatment. The tumor was punctured with a knife and a tube containing radium bromide inserted for three hours at each treatment. Ossification set in, the teeth became firm. Remnants of the giant-cells were found, however, by a pathological examination of the improved portion. The case remains cured at one year from beginning treatment. (Robert Abbe, *Medical Record*, 66, 321, 1904.)

Dancyz<sup>2</sup> demonstrated that the effect is more intense in young than adult animals. He applies this fact to explain the selective action of the rays on neoplasms, while they traverse skin and muscle without appreciable action on them.

1. Henry G. Piffard, *Medical Record*, June 18 (1904).

2. Action du Radium sur les différents tissus, Dancyz. *Semaine Médicale*, Paris, 24, No. 1 (1904).

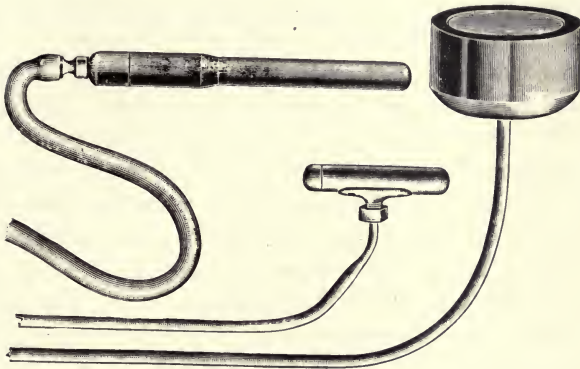


Fig. 61. — Apparatus of Lieber for application of radium compounds in medicine. The tubes are of aluminum. (Through courtesy of Hugo Lieber.)

The envelope of thin aluminum, for reasons already noted according to Lieber, gives greater efficiency than one of glass, mica or quartz. Morton suggests cellulose containers.

• It is assumed, of course, that any physician inaugurating experiments on human subjects will have determined the strength of the preparation before applying it. Even with that knowledge, little is known to-day of the dosage. As adverted to, the pathogenic action, i. e. the destructive effect, evidences itself in temporary hyperaemia or extensive necrosis accompanying a long enduring ulcer. The difficulty in judging this is due to the fact that oftentimes weeks intervene before ulceration becomes apparent. Robert Abbe<sup>1</sup> learned that, as a result of plunging a tube into a mammary tumor, the inactive encapsulation of radium when put into healthy muscular tissue and peritoneum of animals, is no criterion for its action on morbid tissue when buried within the tissues. Upon superficial healthy tissue, radium compounds bring about necrosis by over excitation; upon morbid cells they induce retrograde changes and a substitutive fibro-hyperplasia.

Williams says that under no circumstances should the  $\beta$ - and  $\gamma$  rays be used together for deep-seated diseases, because

1. Loc. cit.

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the  $\beta$ -rays would cause serious injury before the  $\gamma$ -rays had time to produce a beneficial effect.

Einhorn<sup>1</sup> compared the penetrating action of similar preparations of radium with glass, hard rubber, celluloid, aluminum and ivory. Photographic effects indicated that the first three allowed the penetration of the rays better than the last two. He, also, suggested the use of radium in the transillumination of various organs of the body. A capsule of radium was held between the tongue and teeth. The cheekbones became transilluminated. The suggestion was made that the method might perhaps have a diagnostic use in the diseases of the antrum. By the use of his "radio-diaphane" the radium can be carried into the oesophagus, stomach, or rectum. The method of procedure is as follows:

"The patient is examined with an empty stomach; growths from the thorax and abdomen being removed, the radio-diaphane is slightly moistened and introduced into the stomach. A fluoroscope, with barium platino-cyanide screen, is used in observing the rays. All observations must be noted in the dark and after the eyes have become accustomed to the darkness. The apparatus served satisfactorily in determining the position of the large curvature of the stomach; the descending colon or sigmoid flexure, also, may be transilluminated by means of radium, if the radio-diaphane, (Fig. 62), in the bowel is shorter and of stiffer rubber. The bowels should be thoroughly flushed with one or two quarts of water previous to the examination. The instrument is introduced as far as possible without kinking, the patient being placed on his back. The lower abdominal region is inspected by means of a fluorescent screen. It usually requires the inspiration of air to become visible; deep inspiration seems to lessen, while low inspiration increases the luminosity." In transilluminating the lungs from the esophagus, he learned that it was possible to examine them anteriorly and posteriorly.

"Normally, moonshine appears where the lungs are; a faint shadow corresponding to the heart, is observed on the left side. Doubtless marked inflation of the lungs would cause a change in the transillumincancy."

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1. Medical Record, July, 1904, p. 164.

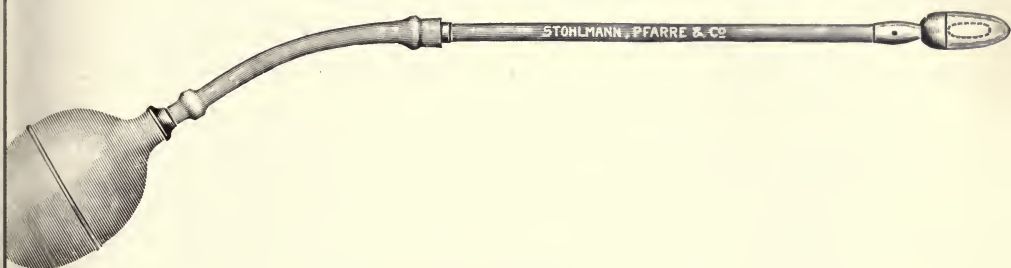


Fig. 62.—The Radiodiaphane.

By the transillumination of the stomach it appears possible to discover tumors, as Einhorn reports he observed in one case. He has, also, treated esophageal cancers in this way with radium. In one case he was able to enlarge a stricture of the esophagus. At first only the smallest size bougie could be passed as far as the lower third; after a month's treatment, however, it improved so that a No. 30 bougie was passed into the stomach and there was no difficulty in swallowing food. From the few cases observed it appeared that partial shrinkage of tumor causes the stricture to be reduced. There were no disagreeable occurrences incidental to the treatment. There was a diminution of pain in some cases, but not in all. No complete cure is reported, but decided improvements were observed.

Exner<sup>1</sup> reported three cases of dilatation of stricture by similar treatment with radium. The stricture resulted from esophageal cancer.

As a further illustration of the variety of evidence and its frequent contradictory character, attention is directed to the statement of Metzenbaum,<sup>2</sup> who says: "From very careful observations no difference could be noticed in the physical or therapeutic results when using radium of 100 activity or 7000 activity."

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1. Wiener klinische Wochenschrift, IV (1904).

2. Louisville Journal of Medicine and Surgery, 188 (1904).



Darier<sup>1</sup> is reported as having used radium successfully as an analgesic and as a curative agent in nervous spasms and paralysis.

Touching the action of X-rays on bacteria, Bear<sup>2</sup> experimented with bacillus coli communus, bacillus typhosus, staphylococcus, streptococcus, Klebs-Loeffer bacillus, etc., using an exposure of one hour at a distance of ten inches, and found no effect, whatever the make of the tube or the method of excitement.

Aschkinass<sup>3</sup> and Caspari<sup>4</sup> first showed that the rays of radium interfere with the development of bacteria. Pfeiffer and Friedberger proved its bactericidal action on saprophytic as well as pathogenic microbes. Dixon and Wigham,<sup>5</sup> continuing their experiments on the action of radium bromide on plants, found in the case of certain bacilli, for example B-pyocyaneus, typhosus, prodigiosus, and anthracis in an agar culture medium, that the  $\beta$ -radiation exercises a marked inhibitory action on their growth. A four-day exposure at a distance of 4.5 c. m. of 5 m. g. of radium bromide does not appear sufficient to kill the bacteria, but arrests their growth, and maintains a patch on an agar plate, inoculated with any of these organisms, sterile. A broth tube, however, inoculated with these in most cases developed the organisms, showing that while the growth was inhibited in the patch, all the organisms were not killed.

Henry Crookes<sup>6</sup> has shown that various bacterial cultures after exposure to the action of 10 mgms. of radium bromide,

1. Consular Report, Guenther, Frankfort, Germany, Mch. 11 (1904).

2. "Effect of Röntgen rays on Certain Bacteria," Journ. Advanced Therapeutics of New York, June (1903).

3. Arch. f. d. ges. Physiol. (Bonn), 86, 603.

4. Allg. Med. Centr. Ztg. (Berlin), 72, 590 (1903).

5. "Action of Radium on Bacteria," Nature.

6. "Bactericidal Properties of the Emanations of Radium," Chem. News, 87, 308.



about 3 cms. distant, were killed. When the plates were incubated for 24 to 48 hours, it was noted that the immediate portion of the plate which had been subjected to the action of the rays showed a bare space free from bacterial growth.

Experiments in our laboratory (University of North Carolina) by Manning, with radium chloride of 7000 activity, indicated an actual stimulation of their growth.

Green<sup>1</sup> found that when bacterial cultures were subjected to the action of radium bromide and then removed, they possessed sufficient activity to affect the photographic plate, even through a double layer of lead foil.

Van Buren and Zinsser<sup>2</sup> report the result of the effect of radium of 300,000 activity upon bacteria. They exposed the bacillus typhus, staphylohemina, pyrogenis aureus from 8 to 19 hours in the dark without any effect. They say this may have been due to the fact that the radium was confined within glass or on account of the shortness of exposure, but they assert that their observations give small promise of achieving brilliant therapeutic results with it as a bactericide, as prophesied by others.

Prescott<sup>3</sup> arrived at the following conclusions :

Radium rays have no effect upon fresh cultures of B-coli, B-diphtheriae, or saccharomyces cerevisiae at a distance of one centimeter when the time of exposure is less than ninety minutes.

Any advantages derived from the therapeutic use of radium must be explained in some other way than by the direct weakening or destruction of the micro-organisms of disease.

The use of radium tubes in the treatment of diphtheria cannot be recommended or regarded as a substitute for anti-toxin.

Ackroyd<sup>4</sup> studied the action of radium on milk, and Schmidt-Nielson reported that the action of radium in the

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1. Nature 70, 69.

2. American Medicine, Dec. 26, 1903.

3. Science, N. S., 20, 247.

4. Nature 70, 55 (1904).

curdling of milk is minimal. Any action it showed upon the chymosin was attributed not to the Becquerel rays, but to the phosphorescence of the generated ultra-violet rays.

— Dr. Margaret Mary Sharpe, of London, appears to have been the first to use radiant matter in the removal of hair as a professional procedure.

From what we have learned there appears to be little in the suggestion that radio-activity may supplant chemicals used for the preservation of food.

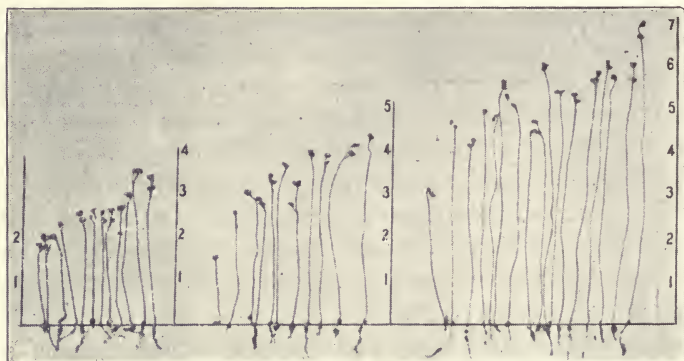


Fig. 63

This shows the hindering effect a radium compound has upon the germination of seed. (Through the courtesy of Dr. Robert Abbe.)

It has even been suggested that radium will solve the problem of determining the sex of children before birth.

Many other suggestions have resulted from the unchecked play of imagination; for example, the prevention of *mal de mer* by the use of radium.

Soddy is reported as having suggested the inhalation of thorium emanations for tuberculosis. Tracy, by a photographic method, reports the radio-activity of the breath after such inhalations.

Soddy has noted that if a radium salt be dissolved in water the emanations are immediately evolved, and collect in



the air above the solution. If the emanations be swept at once into the lungs they serve as a germicidal agent in tuberculosis. Lieber asserts its value in the case of hoarseness with himself.

Morton states he saturated distilled water with radium emanations and this was administered to the patient. It appeared to create fluorescence in the medicines that may have been previously administered. Apparently the rays thrown off from the fluorescing substance become healing agents. This mode of treatment has also been used by Paul-Edward, Radiographer of the General Hospital, London.

Blood has been removed from persons who have acquired radio-activity. It affects photographic plates through translucent substances. This is a case, apparently, of induced activity.

The method, according to Morton, of saturating the water is shown in the accompanying figure. (Fig. 63.)

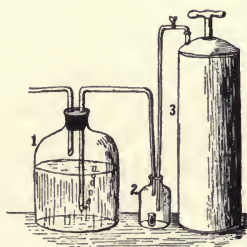


Fig. 63

Morton's method for saturating water with the emanations of radium. The radium compound is in the open vessel in flask 2. Gas is forced by the compressor, 3, over the radium, to sweep the emanations through the water in 1.

Saake<sup>1</sup> refers to the radio-active substances of the air reported by Elster and Geitel as being from 3 to 5 times as great in the mountains as at the level of the sea. "The difference in the tension between the positive air and the negative

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1. "Ein bisher unbekannter Faktor des Höhenklimas," Munchener Med. Wochenschrift 51, 1 (1904).

earth—the potential—also increases with the altitude. Experiments indicate that these electric and radio-active factors have some share in the benefits of the mountain climate and they might be artificially increased.” The writer repeats such statements with trepidation, for all have been either misunderstood or unwarranted conclusions drawn by the zealous newsgatherers with unfortunate consequent delusions on the part of the ill. One instance is reported<sup>1</sup> where at least “one shrewd speculator in human misery proposes soon to start a sort of radium consumption farm, where he will advertise to do wonders for affected lungs by means of radio-active air—and handsome fees.”

Frequent suggestions have been made to prepare salves, ointments, etc., with chemically inert preparations of radio-active substances.

Morton<sup>2</sup> has inaugurated a novel method of treatment by which the introduction of light within the human tissues themselves is claimed. The X-ray and radium compounds are used merely as exciters of the fluorescent substances already within fluids of the human body or by injected fluorescing substances. He says,<sup>3</sup> “I now regard the X-ray and radium as exciters of light, and I think that the curative effects are due to the fluorescent qualities of the fluids of the human body, particularly when these fluids have been made more fluorescent, that is to say, artificially fluorescent by the use of various fluorescent solutions.”

Metzenbaum, however, says: “The conclusions drawn from nearly one hundred experiments give positive proof that while suspending tubes of radium of various strength for long periods in various solutions and various powders, that neither these solutions nor the powders are capable of affecting photographic plates, and are therefore not rendered radio-active, and

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1. “The Sense and Nonsense about Radium,” Cleveland Moffett, Success, April (1904).

2. New York Medical Journal, Feb. 13 and 20 (1904).

3. Personal letter to the writer.

therefore neither the solution nor the powders can in any way affect the metabolism or pathology of living organisms."

In view of the most recent work of Ramsay (Chapter V), it does not seem improbable that substances may become radioactive without actual contact with the emanations.

It is too soon to draw any conclusions from much that has been done. It is unwise to make any final statements. However, we know this much: that the radium rays possess the power of dilating the vessels; that they have an electric action; also, an influence upon the cells of quickly growing tissues and possibly bactericidal properties. These three factors give bright promise of its therapeutic use, when we shall have learned more about this wonderful substance.



## CHAPTER VII.

## OTHER THERAPEUTIC RADIATIONS.

*The Röntgen Rays.*

Attention has already been directed to the fact that when a Crookes tube is placed in series with the poles of a static machine, or the secondary terminals of an induction coil, it becomes the seat of three classes of radiations: (a) the anode rays, or kanalstrahlen of Goldstein; (b) the cathode rays; and (c) the X-rays of Röntgen.

The Goldstein rays are confined to the interior of the tube and hence, from the standpoint of the physician, are negligible. The cathode rays are much more penetrating and in part, according to Oliver Lodge,<sup>1</sup> traverse the tube and possibly may be the chief factor in producing the cutaneous reaction that is observed when the Crookes tube is employed for therapeutic purposes. The X-rays are without the tubes. These are vastly more penetrating than the others. This penetrating power varies inversely with the density of the substance on which they impinge. Substances opaque to light, as aluminum, are readily penetrated, while many substances transparent to light, as rock salt, are remarkably opaque to the X-rays.

It was known for some time that the Röntgen rays, in addition to their value as an aid to surgical diagnosis, possessed peculiar properties which gave promise in the treatment of certain forms of disease, especially those affecting the skin.<sup>2</sup>

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1. Archives of the Röntgen Rays, April (1904).

2. "Lupus," Pusey, Journ. Am. Med. Ass'n., 35, 1476 (1900). The method of Schiff and Freund, of Vienna, was used. In calling attention to the work of Kummel, Pusey states that certainly none of the usual methods of treatment by surgical means could produce such a result.

For a clearer conception of this phase of our subject it becomes necessary to call attention, incidentally, to some of the most recent work in the application of the "X-light" to the treatment of disease. The reader interested in such may secure first hand knowledge of this form of medical practice by referring to fuller and authoritative works.<sup>1</sup>



Fig. 64

The illustration shows a case before and after treatment with X-rays. A. D., twelve years of age. Microscopical diagnosis, lymphosarcoma after first operation and round-celled sarcoma after second operation. Duration, seven years. (Williams, *Medical News*, Feb. 6, 1904.)

Pusey<sup>2</sup> reports the favorable treatment of sarcoma by X-rays, and says that in certain cases, which cannot for any reason be treated successfully by surgical means, the effect of X-rays should be tried. And further, that in cases of sarcoma

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1. As for example, F. H. Williams's, "The Technique of X-ray Therapy as Applied to Diseases of the Skin," and L. E. Schmidt, *Journ. Am. Med. Assn.*, 40, 11, 1903. Also Rollins.

2. *Journ. Am. Med. Assn.*, 38, 166.

which have been treated surgically, the subsequent use of X-ray exposures as a prophylactic, is a procedure which should be considered.

Bartholmy<sup>1</sup> reports a number of cases of cutaneous lesions produced by the application of the X-rays. He urges caution, and states that it is still premature to introduce the radiotherapy in a current practice, one case of burn being observed five minutes after the first application. The physician should not be held responsible for this, any more than for death during chloroform narcosis, when all the rules of science have been complied with. In spite of precautions, accidents are liable to happen when least anticipated.

Rurio-Jicinsky<sup>2</sup> lays down general rules for treatment with X-rays, concerning the kind of tube to be used in protection of the hair, eyes, etc., while Ross and Wilbert<sup>3</sup> found the anaesthetic effect of the X-ray a decided advantage, though they did not find it was valuable as a curative agent in all malignant growths.

Leonard<sup>4</sup> in writing of the Röntgen treatment of malignant diseases, states that the alterative and destructive action produce retrograde changes. In large subcutaneous growths of low vitality, such a rapid destruction may take place as to flood the system with toxins and cause a fatal auto-intoxication and septicemia. The bad effects noted by some observers, such as the stimulation of the growth of tumors, were probably due to this cause, or to under stimulation by too small a dosage. Operative treatment should precede and the X-ray treatment deal with the residue that has escaped the knife. "It must be employed with as great care as any other agent possessing such marked alterative properties."

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1. *Annales de Dermatologie*, Paris, February, 1901.
  2. *N. Y. Med. Journ.*, Nov. 15, 1902.
  3. *Therapeutic Gazette*, Detroit, Feb. 15, 1903.
  4. *Phila. Med. Journ.*, Feb. 14 (1903).

Walker<sup>1</sup> reports the cure of a case of alveolar melanotic sarcoma.

Coley,<sup>2</sup> summarizing the X-ray treatment of malignant tumors, states that they have an inhibitory action on all forms of malignant tumors; yet the number of cases is insufficient to enable us to state what particular varieties are most susceptible to these influences.

Pfahler,<sup>3</sup> in his comments on X-ray treatment of cancer says: "To-day the medical profession seems to recognize it as a valuable therapeutic agent in certain forms of cancer." Among other conclusions drawn, he states that the time required to cure superficial cancer is usually from two to six months. "We can recommend the use of X-ray in all carcinomata, but especially in those that are inoperable or in which operation is refused."

Again this same author<sup>4</sup> gives a number of conclusions drawn from treatment of carcinoma and tuberculosis with Röntgen rays. Among these it may be mentioned that the X-rays are of undoubted value in the treatment of certain cases of both superficial and deep-seated carcinoma and tuberculosis. Yet there are idiosyncrasies in certain persons which render them most susceptible to the X-rays. In these people deeper burns may occur, in spite of the most careful treatment. Epithelioma involving the mucous membrane is much less likely to be involved in these effects than that which involves the skin. There is not likely to be any interference with the sense of sight, even if the X-rays are used directly over the eye. Tuberculosis, whether of the skin or of the glands, yields in certain cases to the X-rays. Epithelioma of the mucous membrane should be removed as soon as possible by the knife and that followed by the X-ray treatment. Operable cases should be operated on and that followed by the X-ray treatment.

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1. Journ. Am. Med. Assn., 40, 1214 (1903).
  2. Med. Record, New York, March 21 (1903).
  3. Journ. Am. Med. Assn., 40, 8.
  4. Journ. Am. Med. Assn., 41, 1406 (1903).

The X-ray is "not only a very valuable therapeutic agent but also a very dangerous one," for as Zeisler<sup>1</sup> has said, "Whoever is making extensive use of the Röntgen rays is bound to have, sooner or later, some unpleasant experience with the much dreaded X-ray burns."

The following diseases have been treated by the X-rays with variable success:

Lupus vulgaris and erythematosus, scrofuloderma, hypertrichosis, acne, sycosis, epithelioma, psoriasis (not permanent), lichenplanus, keratosis palmaris, eczema and pruritus, clavus, hyperidrosis nasi, and dermatitis staphylogenes.

Concerning X-rays and cancer, the editor of the *Journal of the American Medical Association* says, "That the X-rays have a powerful effect on tissue is undeniable. The evidence seems strong, if not altogether conclusive, that they have a selective action on certain morbid celled proliferations; that they check malignant growth by their destructive action on the surrounding healthy tissues is so much less that it can be safely considered as negligible when the beneficial effects are taken into account."

#### *Ultra-Violet Rays.*

The spectrum of the solar rays, as we analyze them at the earth's surface, is found to consist of three distinct portions: (1) At the lower-end certain invisible radiations of comparatively long wave-length, and commonly spoken of as the infra-red portion of the spectrum. So far as we are aware there has been no separate and distinct therapeutic application of these rays, other than their employment as thermic agents. (2) The luminous portion of the spectrum with its colored gamut from red to violet, passing upward from longer to shorter wave lengths from Fraunhofer's lines A to H. (3) The invisible portion of still shorter wave-lengths and indefinite extent, known as the ultra-violet.

Near the surface of the sun this region is undoubtedly of very great length, but as the undulations pass through the

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1. Journ. Am. Med. Assn., 40, 511 (1903).



atmosphere, the waves of shortest length are absorbed and do not reach us. Fortunately these shorter undulations become known to us through artificial sources and it is by this means that physicians have been enabled to utilize the ultra-violet rays in the treatment of disease.

The chief sources of the ultra-violet rays, available for experimental purposes, are the electric arc and the radiations of the spark of the high tension current of a transformer in connection with a condenser.

The electric arc with carbon terminals emits a larger relative proportion of ultra-violet rays than we find in the solar-radiations. If iron terminals be substituted for the carbon, the proportion of ultra-violet is still greater; and if the condenser spark is made to pass between iron terminals, we will have the richest source of ultra-violet rays now known to us.

The most convenient means for detecting the ultra-violet rays are their effect on certain fluorescent minerals, notably willemite and calcite and their ability to ionize gases, as shown by their effect on a negatively charged electroscope.

Willemite associated with calcite and other minerals from Franklin, N. J., and calcite associated with schefferite and braunite from Sweden, serve admirably as aids to an approximate valuation of the ultra-violet rays, as shown by Kunz and the writer.

When subjected to the rays from the carbon-arc willemite fluoresces green, but the calcite is unchanged; to the iron-arc the green fluorescence is more brilliant and the calcite changes from white to very faint pink; when exposed to the rays from the condenser-spark, between iron terminals, the green fluorescence of the willemite is extremely brilliant, the calcite is changed to a bright pink, and in specimens from Sweden to a brilliant red.

When an electroscope, charged negatively, is exposed to the carbon-arc (Finsen-Reyn lamp) it is slowly discharged; that is, in from five to ten minutes; when exposed, at the same

distance, to the iron-arc, in from one and a half to two minutes; and when exposed to the condenser-spark, between iron terminals, in less than half a minute. The discharge of the electroscope in these instances is brought about by the ionizing influence of the ultra-violet rays on the air that lies between the parallel plates of the electroscope. The ionized air thus becomes a conductor of electricity and this permits the charge of the insulated gold-leaf to escape to earth.

The therapeutic utilization of the luminous as well as the non-luminous portions of the spectrum have been thoroughly and well discussed by others, especially on account of the brilliant work of the lamented Finsen.

*The Piffard-Rays.*

Recently there appeared a paper<sup>1</sup> describing what may be termed the Piffard-rays, after the physician who discovered them and who is using them with success in his practice.

The lamp (Fig. 65) is furnished in front with a thin quartz plate, which is transparent to ultra-violet rays, while glass is opaque to them. If the face of the lamp, with the quartz *in situ*, be applied to a piece of photographic paper (Solio) and the lamp actuated by a suitable coil, a strong impression will be made on the paper in about thirty seconds. If the experiment is repeated with the quartz removed, the result is substantially the same.

Ultra-violet rays, as is well known, will discharge an electroscope if charged *negatively* but not if charged *positively*.

On trial Piffard found that the lamp with the quartz in front discharged the *negative* electroscope in about 20 seconds, but with the quartz removed discharged it instantly; that is, within less than one second. He also found further that the radiations from the unobstructed spark would discharge an electroscope charged *positively*.

It was clear from this that in addition to the ultra-violet rays he was dealing with another class of radiations that only

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1. The Medical News, 85, 1057 (1904).

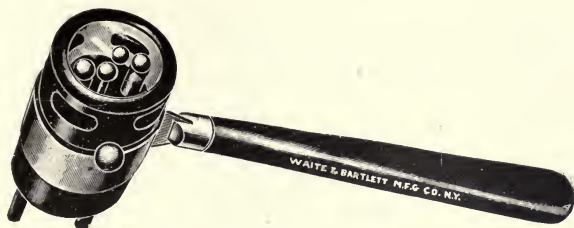


Fig. 65.—Piffard's Ultra-Violet Lamp. The technique recommended is as follows: If the appliance be used with a coil, a single Leyden jar should be employed, with inner armature connected with one of the secondary terminals, and the outer armature with the other terminal of the secondary of the coil. The lamp is then connected directly to the secondary by its cords. Piffard prefers a Wehnelt interrupter adjusted to give a current of five to six amperes through the primary of the coil. The armatures should not exceed 40 square inches of foil in each. This is for the three spark lamp. For the one spark "ionizer" a lesser amount of energy is preferable. The first application should never exceed ten minutes. If connected with a static machine use two Leyden jars, the armatures of which should each have a foil surface of at least 100 square inches. The outer armatures of the jars should be connected together, and the lamp terminals connected to the pole pieces of the static machine. The first application should not exceed 15 minutes with the spark from 15 to 20 millimeters from the lesion.

slightly affected the photographic plate, but acted very energetically on the electroscope.

In default of any means of determining the exact nature of these radiations he assumed that they were negative electrons and predicted that they would act very energetically on the skin or any other tissue with which they came in contact; that the character of the reaction would resemble that from the X-rays and radium, except that it would make its appearance more promptly.

If the radiations in question were negative electrons, as apparently are the cathode rays of the Crookes tube, and the *beta* rays of radium, they would of course be deflected by a strong magnetic field, which Pegram and Milton Franklin found was not the case.

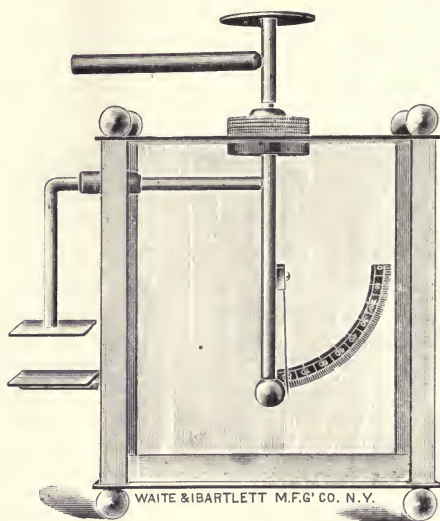


Fig. 66.—Piffard's Electroscopic Instrument, later model.

E. Wiedemann<sup>1</sup> described a new form of radiation to which he gave the name of *Entladungsstrahlen*, and stated that it was not deflectible by the magnet and would not pass through flourspar which readily transmits the ultra-violet rays. He does not appear to have examined the radiations with the electroscopic. It is quite possible therefore that Wiedemann's observations related to the Piffard rays.

In discussing the question of ions, J. J. Thomson says,<sup>1</sup> that if we have a spark one centimeter long in connection with a condenser of 1000 c. m. capacity the pressure developed will be equal to that of 660 atmospheres, (equal to about five tons to the square inch). This pressure, however, diminishes with the distance from the spark according to the law of inverse squares.

When we consider the enormous velocity with which ions are projected in consequence of the pressure behind them, and

1. *Zeitschrift für Electrochemie*, July 20, 1895.

2. *Conduction of Electricity through Gases*. Cambridge, Eng., 1903, p. 392.

the rapidity with which they are developed, it is quite within reason to assume that they will be capable of exerting a considerable influence on tissues that are brought within a centimeter or two of their point of origin. Piffard has found, clinically, that his rays do exert a very powerful influence on the skin; and that the reaction is similar in character to that of the X-rays and of radium; and that it appears much more promptly. Like them also it may produce a curative or a

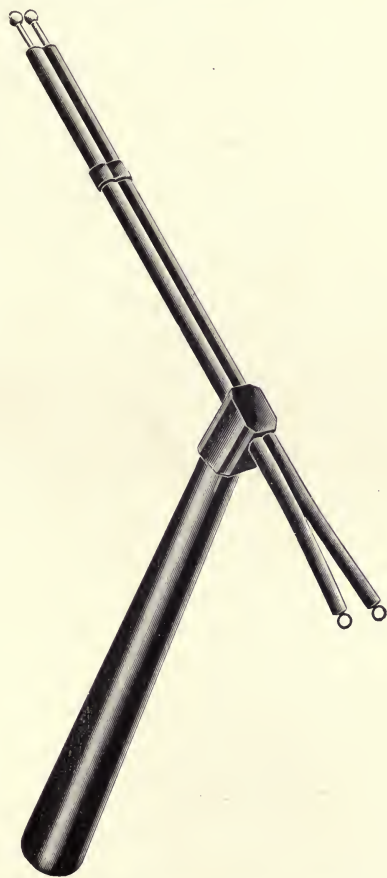


Fig. 67.—Piffard's Spark-ionizer.



destructive effect according to the intensity of the spark and the duration of its application.

When the spark is produced between iron electrodes, with one or more intervening gaps, the total length of the gaps need not exceed one centimeter. If the lamp be used in connection with a coil and suitable condenser, an application of about five minutes with the sparks about 15 m. m. from the tissue, a decided reaction will be obtained in soft morbid epithelial and other degenerating tissue. A similar application for 15 minutes has resulted in the sloughing out of a lupus nodule. It is important, therefore, that care should be used and especially at the beginning of treatment in any given case. These condenser spark radiations have also been used successfully by Robert Abbe, Milton Franklin, and Dieffenbach.

The use of X-rays in connection with uterine cancer and some epitheliomatous conditions of the buccal cavity present mechanical difficulties that it is sometimes inconvenient to overcome. Piffard, to overcome such in the application of his rays, designed what he calls a "spark-ionizer," which may be introduced through a speculum or other suitable shield. The name is given on account of utilization of both the ultra-violet light and the ions (?).

While it is too new for unqualified statements the indications certainly are promising.

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