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RADIATION FROM COMPRESSED POWDERS
OF CRYSTALLINE SUBSTANCES
UNDER HIGH POTENTIALS

A dissertation submitted in partial
fulfillment of the requirements of the degree of

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by

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I. Introduction

In 1922 M. G. Reboul published an article¹ in which he described a photographic effect produced by very small currents passing through a strip of paper. A potential difference, varying between 200 and 1000 volts, was applied to a strip of paper 5 cm. long, and when a photographic plate was placed below, it became slightly darkened. The effect was very faint and required an exposure of from 12 to 48 hours. By employing different absorbing materials placed between the strip and the photographic plate and observing the change in the effect produced on the photographic plate, Reboul attributed its cause to the radiation in the spectrum region between soft X-rays and ultra-violet. As a necessary condition for producing this radiation, the paper must have a great electrical resistance and also have physical discontinuities. He found that the best results were obtained from a strip of filter paper impregnated with a two per cent solution of sulphuric acid and then dried in free air. His investigation was entirely photographic. 171
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In a second article, published a few months later² Reboul employed a pile of several layers of impregnated filter paper placed between a terminal and a metallic gauze. The upper contact was connected to the negative side and the gauze to the positive side of a high tension direct current supply. As a result, the air under the gauze was found to be ionized. This ionization was measured by means of an electrometer and, knowing the intensity of ionization at two different distances from the gauze, Reboul computed the absorption coefficient for air. This value confirmed his previous conclusion that the radiation was in a region between 10 and 500 Angstrom Units dependent upon the applied potential.

In a third paper,³ Reboul introduced a radical change in the construction of the radiating source. Instead of filter paper, use was made of powders of different crystalline substances compressed into the form of a cylinder, Fig. 3 a. The cylinder was placed between a plate A and a gauze B, which were connected to a source of high tension direct current. When a current traversed the cell, the air below the gauze was found to be ionized. He again attributed this ionization to a radiation produced by the cell. By measuring the coefficient of absorption for air and very thin films of celluloid, the range of radiation was located within the same limits as in the previous investigation.

Reboul also found that slow moving electrons and positive particles were projected from the bottom of the cylinder. By employing a method of retarding potentials, the velocity of the electrons was determined. The latter was in good agreement with the values that could be obtained from the Einstein equation

$$Ve = h\nu$$

on substituting for ν the frequency of the emitted radiation as determined from the values of the absorption coefficients for air or celluloid.

This led Reboul to the explanation of the observed phenomena. According to his theory, the radiation is caused by the electronic impacts taking place in the surface layer of the material near the bottom of the cell. Due to discontinuities in the structure of the cell, a potential drop may be established in this surface layer and the electrons accelerated through it. Some of the moving electrons lose their energy on colliding with the molecules of the material substance, thus causing an emission of radiation.

Seven years have passed since the publication of the first paper by Reboul, and not a single paper on the subject has appeared since then in the current scientific literature by anybody else. The importance of the work of Reboul lies in the fact that it provides a new method for obtaining radiation in an important gap connecting X-rays with ultra-violet. It is the object of this dissertation to describe the experiments which were performed in order to determine whether the effect observed by Reboul could be reproduced. During the course of experimentation it was found that in order to make a radiating cell which will produce very absorbable radiation, a number of factors which were not mentioned by Reboul must be carefully controlled. These essential conditions together with the experimental technique and the manner in which they control the character of the emission from the compressed powders of crystalline substances will be described in what is to follow.

II. Experimental Arrangements

The diagram of connections, Fig. 1, represents the apparatus employed in this research. A 110-volt alternating current line was connected across an auto-transformer which fed the primary of a 20000-volt Aome transformer. The secondary of this transformer was connected through a 150000 Ω protecting resistance to a Kenotron rectifying tube and to the testing box containing the radiating cells. On account of only half-wave rectification, eight condensers each of 0.0017 M. F. capacity were connected in parallel in order to smooth out the rectified wave form. These condensers were protected by a spark gap adjusted for 15000 volts.

The testing box was connected as indicated in the diagram. A galvanometer was placed on the grounded side of the circuit and measured the current passing through the testing box without being affected by any leakage currents through the electrostatic voltmeter or condensers. A variable shunt resistance was connected across the galvanometer so that its sensitivity could be varied from 2×10^{-8} to 3×10^{-6} amperes per division.

An electrostatic voltmeter, with a range from 2000 to 10000 volts, was used to measure the potential applied to the radiating cells. In order to be able to measure accurately a potential between 500 and 2000 volts, the auto-transformer was calibrated. This was done by introducing a resistance of ten megohms in place of the specimen and then regulating the auto-transformer until the value of the current through the galvanometer corresponded to 500, 1000, or 1500 volts. These points were marked on

DIAGRAM OF CONNECTIONS

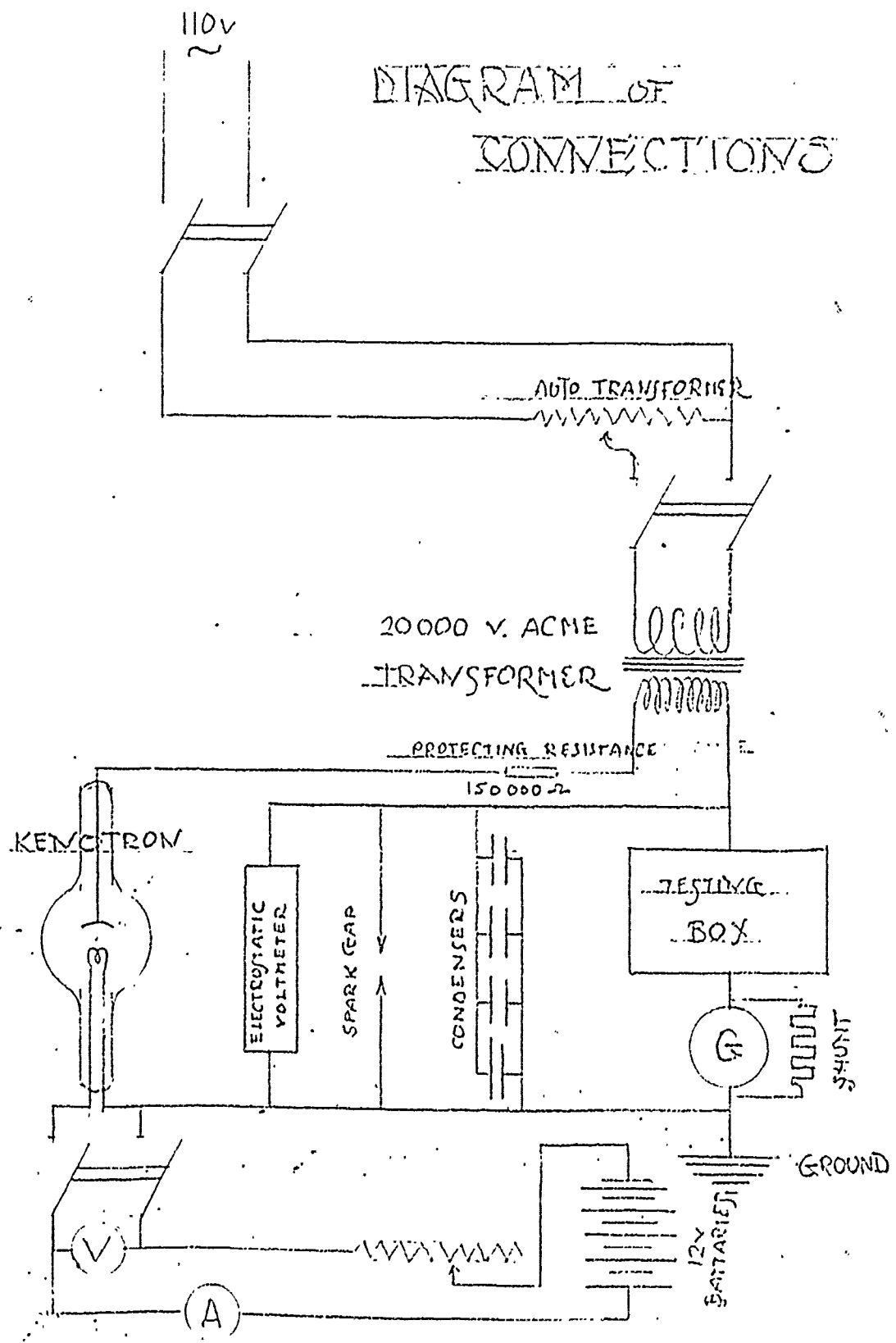


FIGURE 1

the auto-transformer and used as reference points for potentials less than 2000 volts. Incidentally, by the use of this method the readings of the electrostatic voltmeter were checked and found to agree within five per cent in the range between 2000 and 10000 volts.

A photograph of the assembled apparatus is shown in Fig. 2.

III. The Radiating Cell and its Accessories

The description of the radiating cell used by Reboul is limited to a few lines: "La cellule à rayonnement comprend, en principe, une pastille d'un sel qui a été d'abord finement pulvérisé, puis aggloméré à la presse à pastiller." If such a block of compressed crystalline powder be placed between two electrodes, the lower of which is a metallic gauze, upon the application of potential, a large number of minute arcs or sparks can be observed at places where the gauze does not make a good contact with the compressed powder. It was then tried to compress the powder directly on a gauze. While this method decreased the arcing considerably, the contact between the gauze and the crystalline substance did not remain permanent.

The results of Reboul, as far as the ionization of the air below the gauze is concerned, must be regarded with grave doubt. Such ionization is more likely to have been produced by arcing than by a radiation emitted from the cell. This explanation may account for the very large absorption coefficient obtained by him in the case of thin sheets of celluloid. The reduction in ionization might have been caused, not so much by absorption of the radiation which supposedly caused ionization, as by the screening effect produced by the film of celluloid which evidently could not be penetrated by the ions produced at the localities of sparking.

This objection makes it imperative to construct a radiating cell in such a manner as to eliminate any arcing at the junction of the lower electrode and the crystalline powder. The problem presented more

vexatious difficulties than were anticipated. More than twenty different kinds of cells were tried, leading finally to a construction which in every way proved to be satisfactory.

The final form of the radiating cell is represented in Fig. 3. It is essentially composed of four parts: (1) an outer shell, (2) a compressed crystalline powder, (3) an upper terminal, and (4) a lower terminal.

An outer shell was found to be essential, due to the fact that, under low pressures, the cohesion between the grains of the powder was not sufficient to bind the whole mass into one solid block. A fibre tube, 1.6 cm. inside diameter and 2.0 cm. long, was used for this purpose. Since this tube would not withstand pressures above 50 Kg/cm² it was necessary to insert it into a strong steel ring during the compression.

A lower terminal was made of a sheet of tin foil, 0.01 mm. thick. The central part of the foil had a hole 11 mm. in diameter, with 12 smaller holes punched around it. Some powder would be compressed into these small holes and thus provide an intimate contact with the crystalline substance.

The upper terminal consisted of a circular disc of bakelite which accurately fitted into the fibre tube. Through the centre of this disc passed a screw with a little plunger. The latter was pushed against the compressed powder by a small helical spring located inside the screw.

The preparation of the cell consisted of the following operations. The material to be investigated was powdered in a mortar, then passed through a silk sieve and retained on another silk sieve of a finer mesh. In this manner it was possible to obtain uniform grains

RADIATING CELL

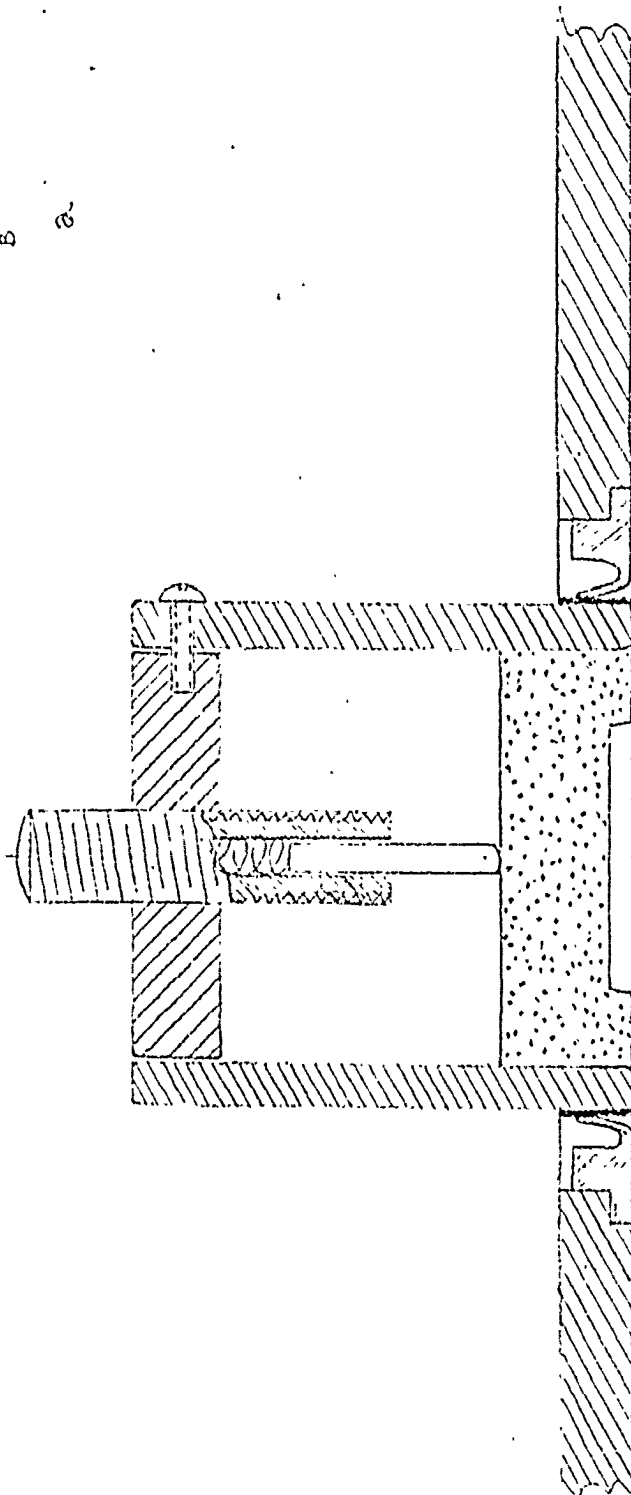
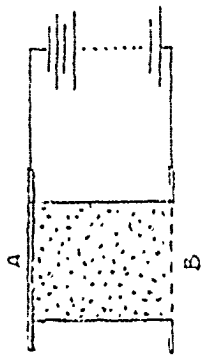


FIGURE 5

ranging between the upper and the lower limits of the two sieves employed. The amount of powder used in all the experiments was one gram. The fibre tube was forced into the steel ring and the lower terminal attached to the fibre tube by putting some glue on the lower edge of the tube. The ring with the tube and the terminal was placed then on a steel base, the central part of which was elevated so that after compression it left an indentation in the middle portion of the bottom as shown in Fig. 3. The powder was then put into the tube and a steel plunger forced down until the required pressure was obtained.

After compression, the fibre tube was forced out of the ring. The tin foil terminal was bent around the edge of the fibre tube to provide contacts when the latter was inserted into a brass ring in the cell holder. The upper terminal was then attached to the tube and in this form the cell was ready to be placed in the light-tight box for the investigation of its characteristic radiation. This investigation was done by photographic means. The radiation from the cell was recorded on a photographic film located directly under the bottom of the emitting surface. At the beginning only one cell was employed at a time. It was found, however, that there existed a large number of factors which determined the character of radiation. In order to be able to understand these factors, investigate their influence and evaluate their importance, resort was made to the method most commonly employed in the investigation of any complex phenomenon. Six cells were constructed under identical conditions with the exception of one factor which was assumed to affect the nature of the radiation. This one factor

was made to vary in steps from one cell to the other. By applying the same potential to all of these six cells and examining the final effect, it was possible to draw a conclusion as to the importance of that one variable.

This led to the construction of a light-tight box in which six radiating cells could be investigated simultaneously. The general view of the box, with the six radiating cells and the film holders in place is given in the photograph, Fig. 4. The box was constructed of bakelite lined with black velvet in order to eliminate the possibility of any spurious effects due to the reflections from the walls. Its dimensions were 21 cm. x 27 cm. x 11 cm.

The cell holder was made of bakelite with six brass rings inserted. These rings were made as illustrated in Fig. 2. A piece of wire connected each ring with a separate terminal at the bottom of the box, leading to a switch, as indicated on Fig. 5. By throwing a switch into the position 1 that particular cell could be connected directly to earth, whereas if the switch were thrown into position 2 it was connected to earth through the galvanometer. In this manner it was possible to measure the current through each cell individually without disconnecting the others. The bakelite plate with the six cells could be removed from the box by slipping it out of two parallel guides; the cell holder could also be adjusted in a vertical direction or turned through an angle.

The film holders were made of brass plates and were provided with a string which, passing around every corner held the photographic film as well as the absorbing sheet of celluloid or cellophane. The arrangement by means of which this was accomplished is represented in

Fig. 6. A small cylinder which fitted accurately into a brass tube was soldered to the other side of each of the brass plates. The film and the absorber could be attached to the plate and the latter mounted in the tube which passed through the double walls of the light-tight box. This tube could be slipped in and out, thus providing a means of adjusting the film to any desired distance from the bottom of the radiating cell.

In order to eliminate any electrical discharges between the bottom of the cells and the film holders, the latter were also connected to the ground. This arrangement probably affected the distribution of the electrical field inside the cell. The importance of this point will be considered in connection with the mechanism of radiation.

IV. Spectra of the Visible Radiation

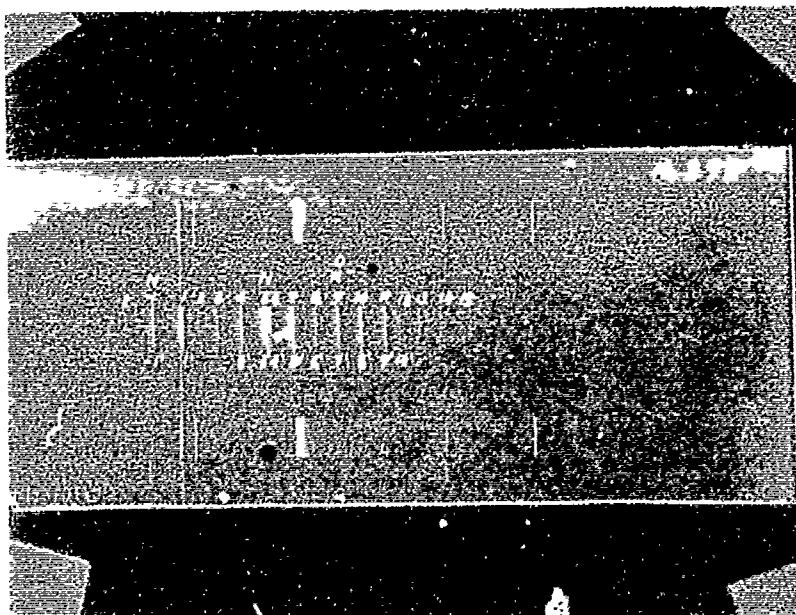
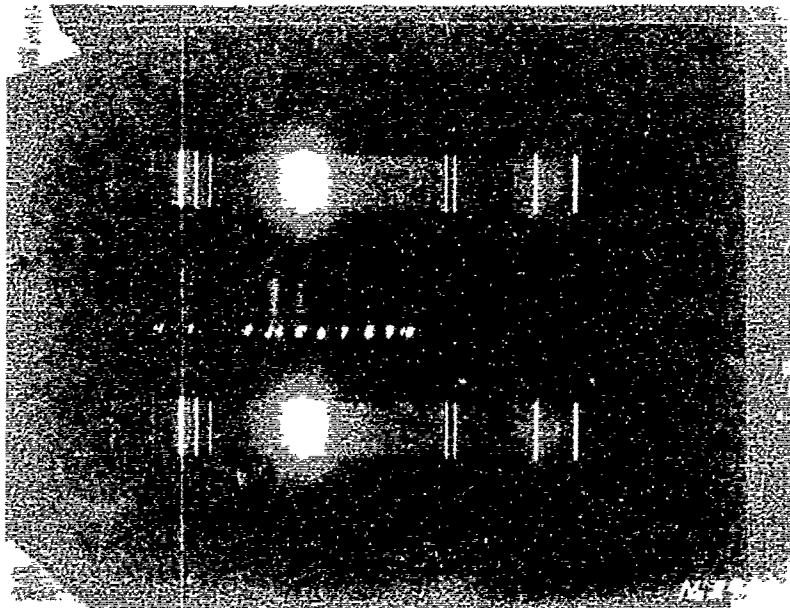
The first radiating cells were made of alum crystals pulverized in a mortar. The grain size was not uniform varying probably between .0025" and .03". The pressure applied to these cells was not measured accurately, it certainly did not exceed 75.Kg/cm². At a potential of about 4000 volts the bottom of the cell began to emit visible radiation of a bluish-green tint which passed without any absorption through a piece of glass one millimeter thick. This radiation was analyzed by means of a Baush and Lomb Constant Deviation Spectrograph. Due to the fact that the intensity of radiation was very small, the cell had to be placed very near to a wide slit and the duration of exposure had to be long.

The cell was mounted in a light-tight box attached directly to the collimating tube of the spectrograph. Fig. 7 represents the spectrum obtained from the radiating cell with an applied potential difference of 8000 volts, the current varying from 5 to 1 μ a the width of the slit being .2mm. and the time of exposure 13 hours. For the purpose of comparison, a mercury arc spectrum was photographed on the same plate. The spectrogram obtained showed eleven lines in the visible part of the spectrum. Next it was necessary to obtain the spark spectrum of alum and compare the two. For this purpose two electrodes were prepared according to the sketch in Fig. 8. The outer shell for each electrode was made out of a bakelite rod turned on the lathe to form a cup with a hole running through its bottom. A piece of copper wire was inserted first and then some alum powder was compressed into the cup. The two electrodes were separated by a distance of .2mm and, when a potential difference of 8000

volts was applied to them, a current between 6 and $3 \mu a$ was passed continuously. The electrodes were set up in the same manner as the radiating cell in front of a slit .03 mm wide and the plate was exposed for 13 hours. Again for the purpose of comparison a mercury arc spectrum was obtained on the same plate. Knowing the wave-lengths of the mercury arc lines, the calibration curve for the spectrograph was constructed. The zero line on both spectrograms was chosen as the mercury line 4046 angstrom units. In Fig. 7 the lines were marked from -1 to 10 and in Fig. 9 from -2 to 15. The lines in Fig. 9 which corresponded to the lines in Fig. 7 were marked by the same numbers primed. There existed a complete identity between these two series of lines as far as their wave-lengths were concerned. This explained the visible radiation emitted by the compressed powder of alum, the effect being due to the sparks or arcs which occurred within the cell upon the application of a high potential difference.

The conductivity of alum crystals is given by Joffe as $12 \times 10^{-16} \text{ cm}^3/\Omega$, ⁴ while the conductivity of compressed alum powder was found to be only $5 \times 10^{-11} \text{ cm}^3/\Omega$. This tremendous increase in conductivity was probably due to the moisture which was adsorbed by the surfaces of the little crystals of alum. The conduction evidently takes place over their surfaces. In a non-homogeneous substance we have to consider the resistance as being made up of an arrangement of resistances and capacities in series and in parallel. The structure of such a substance is not continuous and if a potential is built to a sufficiently high value a spark discharge will occur at the place of discontinuity, accompanied by a corresponding ionization. The transfer of charges

across such a heterogeneous conductor is due to both polarization and conductivity. The spectrum of the characteristic radiation of such a conductor, i.e., radiating cell, corresponds to a spectrum produced by a spark discharge between two electrodes composed of the same substance as that of the cell.



V. Absorbable Radiation

The presence of visible radiation emitted by an alum cell did not exclude the possibility that under other circumstances the compressed alum powder would emit very absorbable radiation. In fact some of the cells which were made showed the presence of such radiation, but the results could not be reproduced at will. It was clear that there were several factors which determined the character of radiation from compressed crystalline powders and it was the purpose of further investigation to determine the exact conditions under which absorbable radiation could be produced.

Test No. 420

In the first place the effect due to different pressures, under which the crystalline powder of alum was compressed, was determined. One gram of alum powder with grain size of .0025" was put into fibre tubes and a pressure varying from 57 Kg/cm² to 2250 Kg/cm² was applied to them. These cells were placed in the light-tight box and a potential difference of 5000 volts applied to them. The upper half of the photographic film was covered with a piece of celluloid 0.13 mm thick. If there were any absorbable radiation emitted by the cell, the upper part of the film covered by celluloid ought not to show any darkening. The developed films showed that most of the radiation was limited to the visible part of the spectrum and its intensity increased very regularly with the pressure under which the cells were made. Film No. 420, a and b, showed a considerably more absorbable radiation than the rest of them.

No 420

No 420

a

b

c

d

No 420

e



No 421

No 421

a

b

c

d

No 421

e



No 422

No 422

a

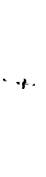
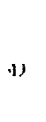
b

c

d

No 422

e



Tests Nos. 421 and 422

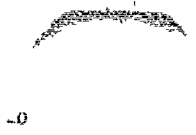
Two more runs were carried on six radiating cells No. 420. The low pressure cells ceased to show the presence of absorbable radiation. This was due to the fatigue mentioned by Reboul and was substantiated also in some of the later experiments. It is a peculiar fact that the absorbable radiation which may be recorded during the first twelve hours of exposure disappears almost completely in successive exposures, although the current through the cell may be increased two or three-fold.

The increase in the intensity of radiation with the increase in pressure can be attributed to the fact that under greater pressure the compressed alum powder becomes translucent. Under this condition any light coming from the interior of the cell will be transmitted. We may consider, therefore, that the intensity of radiation is a function of the degree to which the substance of the cell is rendered translucent. It was thought at first that under high pressures the possibility of sparking in the interior of the cell would be decreased and therefore the radiation, if any, might be of the more absorbable type. This conclusion was not supported by the experiments, at least up to a pressure of 2250 Kg/cm².

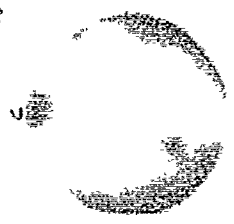
Test No. 423

As stated before, when alum crystals were pulverized, the conductivity of the compressed powder increased considerably. It was the object in Test No. 423 to find out to what extent the humidity of the air affected the resistance of the cell and the character of the emitted radiation. In the previous test the relative humidity of the air was

a N-423



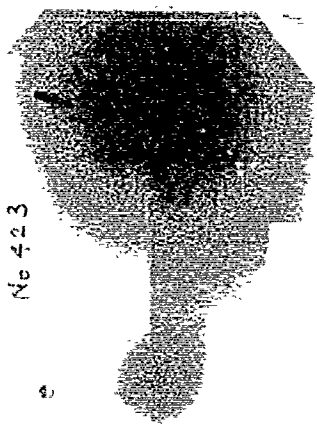
b N-423



c N-423



d N-423



a N-425

c

b N-425

d

r

65 per cent, while in the present test it was only 55 per cent. The pressure applied to the cells varied from 23 Kg/cm² to 2250 Kg/cm².

A difference of ten per cent in humidity caused a five-fold decrease in conductivity. The effect of humidity on conductivity supports the explanation that the specific resistance of compressed crystalline powders depends upon the amount of moisture adsorbed by the powder.

The developed films showed again that the intensity of radiation increased with the increase in pressure.

Film No. 423 a and b indicates the presence of radiation which was completely absorbed by celluloid 0.13 mm thick. The moisture content of the powder was thus a very important factor determining one of the conditions under which a cell emits only absorbable radiation.

From the photographic studies it could not be decided whether the absorbable radiation was also included in the visible radiation emitted by highly compressed cells. The intensity was far too great to permit the observation of any difference due to the absorption that might have been caused by the celluloid. It can be stated, however, that if such radiation were present, its intensity was many times smaller than the intensity of the visible radiation.

From the above tests two important factors were found which determined the conditions for producing very absorbable radiation:

(1) The amount of moisture adsorbed by the powder, (2) The pressure under which the crystalline powder was compressed into the fibre tube.

Test No. 425

The object of this test was to find out whether the conditions

as determined in the previous two tests were sufficient to provide reproducible results. Six radiating cells were made out of alum powder similarly to No. 423 a and b.

The developed films show that the character of the radiation is the same as that observed previously.

Tests Nos. 428, 429, 430

In all the previous tests the size of the grains of the crystalline powder of alum was .0025". The object of the present tests was to determine what relation existed between the size of the grains, the applied potential difference and the character of the emitted radiation. In the preparation of the cells the constancy of the two factors determined before was maintained, i.e., the relative humidity of the air and the applied pressure. The new variable introduced into Test No. 428 was the size of the grains, which ranged from .0296" to .0025". The developed films No. 428 indicate that absorbable radiation is emitted only when the size of the grains does not exceed .0032".

Test No. 429, carried out on the same set of radiating cells, showed that when the potential decreased to 3000 volts absorbable radiation was emitted only by the cells the size of whose grains did not exceed .0057".

Finally, Test No. 430 showed that when the applied potential was reduced to 2000 volts a cell with still larger grains changed its radiation from visible to absorbable.

As a result of these tests two more factors were discovered which determine the conditions under which a compressed crystalline substance becomes an emitter of only absorbable radiation: (1) the

size of the grains; (2) the applied potential.

Test No. 432

In previous discussions the term "radiation" was freely used in describing the cause of the darkening produced on the photographic film placed under the bottom of the cell. It was natural to raise the question, whether the same effect could not be attributed to chemical action or the mechanical action produced by the bombardment of the film with electrons or ions that are probably ejected from the bottom of the cell. If the effect is caused by radiation, then by employing absorbers of different thicknesses the darkening of the photographic film will be shown to be a function of the thickness. This was the object of Test No. 432. As absorbers a piece of celluloid 0.13 mm thick and a piece of cellophane .02 mm thick were used.

The developed film No. 432 e and f indicates that some of the radiation passed through the cellophane while it was completely absorbed by the celluloid. If the effect were due to ozone, no darkening would be observed under the cellophane which is impervious to a gas. Since neither an electron nor an ion would pass through a film of cellophane 0.02 mm thick, it was proper to attribute to radiation the darkening of the photographic films observed in all the previous tests.

Tests Nos. 434 and 435

According to Rebul the property of compressed crystalline substances to emit very absorbable radiation was not limited to alum alone. A test was carried out also on some other substances such as coal, lead acetate, sulphur, sodium chloride, and copper sulphate.

Films Nos. 434 and 435 show the effect produced by each of the mentioned substances under potentials of 2000 and 3000 volts.

N. 432

a



b



N. 432

c

d

e

N. 432

f



N. 432

g

h

N. 432

i

j

k

l

N. 432

m

n

N. 435

o

p

N. 435

q



r

s

N. 435

t



u

v

w

x

y

z

VI. The Mechanism of Radiation

VII. Conclusions

The results of this investigation confirm the conclusions of Reboul that under proper conditions, compressed powders of some crystalline substances become sources of very absorbable radiation.

The investigation led to the construction of a radiating cell in which the contact between the electrode and the crystalline substance was made in such a manner as to eliminate entirely the possibility of sparking. Such a cell, when made under specified conditions gives reproducible results.

The factors regulating the characteristic radiation from compressed crystalline substances are:

1. Chemical composition
2. Applied potential
3. Relative humidity of the air
4. Size of the crystalline grains
5. Pressure

The necessary conditions under which an alum cell emits absorbable radiation can be summarized as follows. For a potential difference of 5000 volts, the powder must be prepared and compressed in an atmosphere whose humidity is less than 55 per cent. The average size of the grains must be less than .0032", and the pressure under 300 Kg/cm².

Under the above conditions the emission from such a cell is completely absorbed by celluloid $t = .15$ mm and only partially absorbed by cellophane $t = .02$ mm. This indicates that the range of the radiation lies between the extreme ultra-violet region and very

soft X-rays.

In closing the author desires to express his deepest gratitude to Dr. L. T. More and Dr. R. C. Gowdy whose kind interest has made this research possible. Acknowledgement is also due for the assistance received from other members of the Physics Department of the University of Cincinnati.

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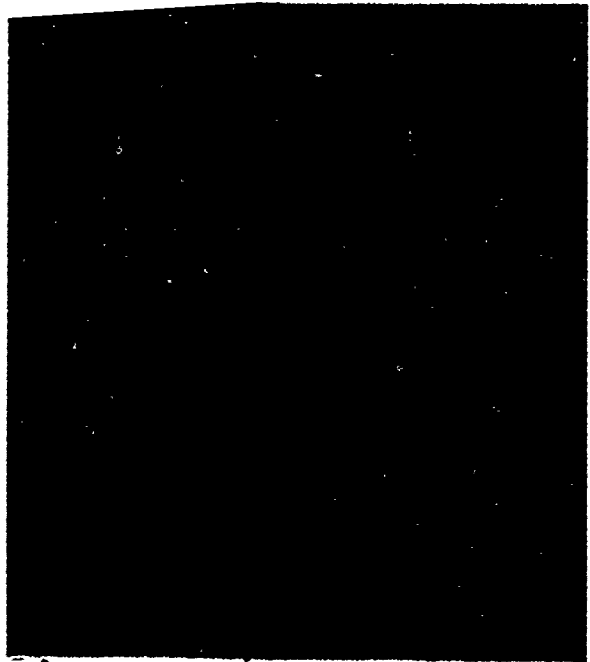
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