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THE LUMINESCENCE OF PURE BARIUM BROMIDE UNDER THE ACTION OF ALPHA-, BETA-, AND GAMMA-RAYS

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Abstract

The luminescent material studied (c. p. barium bromide heat-treated) is not luminous by itself, nor is the exciting agent (radium sulphate covered with varnish) self-luminous under the existing conditions. By the use of an especially designed brass capsule the barium bromide was held within the range of action of the α-,β-, and γ-rays from the radium, but the luminescent material and the exciting agent were kept separate. Under these conditions there was for a time, a definite increase in brightness, then from maximum brightness the luminescence decayed with time until a limiting value was reached. Samples of barium bromide from three manufacturers were studied, each under the action of four different amounts of radium. The results were consistent. It was found that the the initial luminosity varies with the temperature at which the barium bromide has been treated previous to the action of the rays. The sample was heated to the desired temperature, then cooled quickly to room temperature, pulverized, placed in the capsule in contact with the radioactive preparation and the brightness measured as soon as possible. The luminosity gradually increased with the temperature of heat-treatment to a maximum value, then the luminescence decreased and rose again to a second maximum, which was followed by a gradual decrease with further increase in the temperature of the heat-treatment through the melting point of the barium bromide.

Introduction

THE decay of luminescence of zinc sulphide due to radium rays has been investigated by several experimenters,[1] and theories have been formulated to explain their results. In the case of barium bromide the initial growth of the luminescence and the variation with the temperature of previous heat-treatment are difficult to explain by these theories.

Przibram[2] and his students while studying photoluminescence in kunzite and other minerals noticed a time variation of the brightness. They found that the luminescence of some minerals increased under the action of β-rays and then gradually decayed after passing through a

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[1] E. Rutherford, Proc. Roy. Soc. **83A,** 561 (1909–10); E. Marsden, Proc. Roy. Soc. **83A,** 548 (1909–10); Paterson, Walsh and Higgins, Proc. Phys. Soc. of London **4,** 215 (1917); J. Ewles, Phil. Mag. **45,** 957 (1923).

[2] K. Przibram and E. Kara-Michailova, Akad. Wiss. Wien, Ber. **131,** 2A 285 (1923); **132,** 2A 261 (1924).

maximum. Rodman[3] showed the increase of brightness with the rise in temperature of heat-treatment in the case of pure radium bromide. There appeared one maximum at about 600°C. Karrer and Kabakjian[4] found a variation with temperature as they heated luminescent radium compounds.

Studies in luminescence, up to this time, have been made using mixtures of the luminescent material and the radioactive exciting agent, or the effects have been produced by the use of x-rays, or cathode rays. One of the objects set for this experiment was to keep the source of radiation separate from the luminous material, in order to avoid the formation of any possible complex products. In addition it was thought desirable to use a source of rays that would remain constant through long intervals of time. Under these conditions an attempt was made to investigate the variations with time, and with heat-treatment at different temperatures, of the excited luminescence in pure barium bromide using a deposit of radium salt as the source of the α-, β-, and γ-rays.

METHOD AND APPARATUS

An old type Nutting polarization photometer, made by Hilger, was used. The optical system may be represented by the diagram Fig. 1.

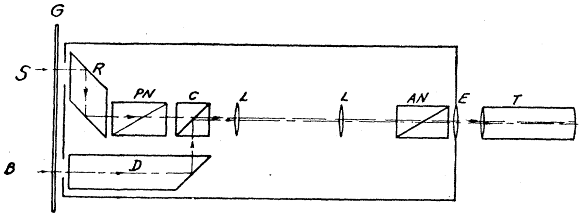

Fig. 1. Diagram of photometer: *S*, standard lamp; *G*, ground glass screen; *R*, reflecting rhomb; *PN* and *AN*, nicol prisms; *C*, photometer cube; *L*, *L*, *E*, lenses; *T*, telescope; *B*, luminescent sample; *D*, reflecting prism.

The standard lamp used was a 110-volt incandescent carbon filament lamp burned at 60 volts. This in turn was compared with two similar lamps calibrated at the Bureau of Standards. Constant lamp voltage was attained by the use of a potentiometer arrangement in a circuit using storage batteries as a source, and a ballast lamp to stabilize the current for small variations in the battery voltage.

The chief difference from other experiments in luminosity lies in the method used to produce the luminescence and at the same time to keep

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[3] Miss J. A. Rodman, Phys. Rev. **23**, 478 (1924).

[4] E. Karrer and D. H. Kabakjian, Jour. Frank. Inst. **186**, 317 (1918).

the materials pure and separate. Brass capsules were prepared as shown in Fig. 2, each consisting of four parts. On the face of the thimble (4) was soldered a gold plate upon which was spread a thin layer of radium sulphate. The purpose of the gold plate was to facilitate the recovery of the radium after its use. This layer of radioactive material was covered with a coat of varnish. One coat was usually enough to keep most of the decay products of the radium sulphate underneath and still be thin enough to permit the α-rays as well as the β- and γ-rays to pass through. The radioactivity was tested from time to time and found to reach an equilibrium value which has remained constant for a year. The sleeve (3) served to hold the parts (1) and (4) together. A lead washer (2) provided a means of holding a small amount of barium bromide of a definite thickness in the proper place between the radioactive material and the window in the brass cap (1). A piece of very thin mica covered the luminescent sample and the lead washer. Against this mica disc the cap (1) was screwed snugly. This arrangement was easy to use, could be kept in a desiccator when not in use, provided a definite thickness of the sample and held it in place. By a series of trials a thickness of sample was found which produced maximum brightness. This thickness, which is about the range of the α-particle in the powdered barium bromide, was then used with exciting sources of 0.3 to 0.9 milligrams of radium in the form of radium sulphate spread over an area of about one square centimeter.

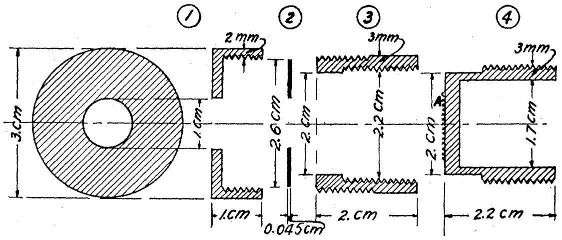

Fig. 2. Brass capsule. 1, a cap; 2, a lead washer; 3, sleeve; 4, thimble; *A*, the radioactive material.

The barium bromide used in these measurements was heated to various temperatures in a hard glass or a quartz tube, allowed to cool quickly to room temperature, then ground in an agate mortar and placed in the capsule in powder form. All measurements were made at room temperature.

The errors involved are those due to personal error in reading very low intensities of light, and those due to the optical system employed.

RESULTS

This study has shown for a pure substance (c.p. barium bromide) what has been observed in mixtures; first, the growth of the brightness to a maximum value as time progresses, followed by the decay of the luminescence with time from this maximum to a constant value; second, the variation of the initial luminosity with the temperature to which the sample has been subjected before exposure to the radium rays.

The luminescence as shown in Fig. 3a gradually grows to a maximum value under the continuous action of the constant source of α-, β- and γ-rays. From this maximum the luminosity gradually decays to a constant value under the action of the same source of rays. The time to reach the maximum brightness varies with the amount of radium present. In Fig. 3a, the barium bromide used was from the same maker, but the four capsules used carried different amounts of radium in them. Number 1 with the least amount of radium takes the longest time to reach its maximum, and this maximum value is not so great as the maxima in which stronger bombarding sources are used. This shows that the strength of the α-, β- and γ-ray source affects both the brightness of the sample and the time required to reach its maximum luminosity. As the curves are all plotted on the same scale it is seen that the stronger the radioactive source the brighter is the sample throughout the course of the study and the sooner it reaches its maximum value. In curve No. 1 Fig. 3a, the maximum is attained in about twenty hours, with 0.293 milligrams of radium present. In curve No. 2, the time for this maximum is about fourteen hours, with 0.522 milligrams of radium on capsule No. 2. In curves Nos. 3 and 4 in Fig. 3a, where the time required to attain the maximum luminosity is about nine hours, sources of 0.910 and 0.815 milligrams of radium were used on capsules 3 and 4 respectively.

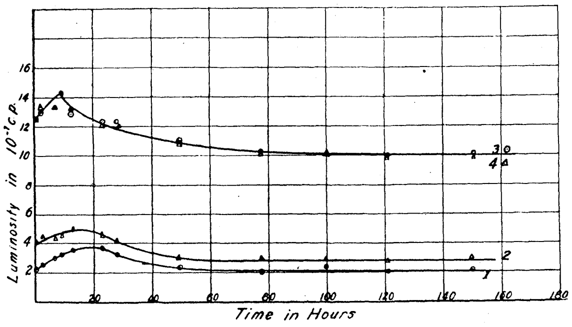

Fig. 3a. Time-luminosity variation. Barium bromide prepared by Mallinckrodt was used for these curves. The number of the curve corresponds to the number of the capsule.

Fig. 3b shows the time-luminosity study using the same capsule (No. 4 in this case) with three different preparations of supposedly pure barium-bromide. Samples from Merck, Mallinckrodt and Kahlbaum show the same general characteristics as to the shape of the curves and the time of reaching the maximum, although the actual values of the brightness at the maximum and the rate of the decay vary somewhat in the different samples. An attempt to purify one of these samples, that from Kahlbaum, by five successive recrystallizations from c.p. hydrobromic acid (Merck) failed to produce any definite change in the luminescence.

A possible explanation of the rise in the luminescence might be assigned to small amounts of the emanation leaking through the varnish and producing luminescence by an active deposit on the salt itself. If this were true, the compound should show considerable luminescence when the source of radiation is removed. This was tested by removing the sample from the capsule and examining it in the dark, but no luminescence comparable to the rise observed in these experiments could be detected. A very feeble luminescence was noticed, which probably was due to a slight degree of thermoluminescence in the material. This effect completely disappeared on heating to about 200°C, showing it could not be due to an active deposit from the radium emanation. As further evidence that escaped emanation cannot be the cause of the effects noticed, the lid of the capsule containing the barium bromide was removed and the compound exposed in a desiccator for a time long enough to allow any emanation that it might contain to diffuse away. On replacing the cap the luminescence had not measurably changed, showing that the growth of the luminescence cannot be explained on this basis.

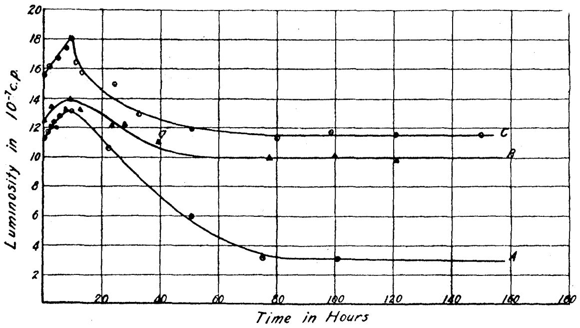

Fig. 3b. Time-luminosity variation. Curve *A*, Kahlbaum barium bromide used in capsule No. 4; *B*, Mallinckrodt barium bromide in No. 4; and curve *C*, barium bromide from Merck in the same capsule, No. 4.

Long-continued exposure to the radiation causes the barium bromide to acquire a brownish color. This phenomenon has been observed by numerous experimenters before and was noticed in this case also. This brownish-colored barium bromide shows thermoluminescence.

The second result of this study is the change of the luminescence produced by changing the temperature to which the sample is subjected before measurement. The sample was brought to the desired temperature which was measured by a Thwing thermoelectric pyrometer, then removed from the furnace, cooled to room temperature, put into the capsule and the initial luminescence measured. Ordinary unheated pure barium bromide does not show appreciable luminescence under the action of the α-, β- and γ-rays. After heating at 100°C the luminescence which is excited by the sources used is barely measurable. With slightly higher temperature the barium bromide gives up its water of crystallization, with no sudden change in the luminosity. No progressive change in the initial luminosity could be detected by continuous heating at a constant temperature of 290°C for a week. With higher temperatures the luminescence rises gradually until it reaches a maximum, for Mallinckrodt material at about 650°C. More intense heating results in a sudden drop in the brightness. This decrease is followed by a sharp rise in the luminescence until a second maximum is reached, at about 750°C for Mallinckrodt material. From this point the initial luminescence falls off at least until the melting point is passed. There seems to be no sudden change in the luminescence due to fusion during the heat-treatment. Fig. 4 shows the results of such a temperature study of luminescence

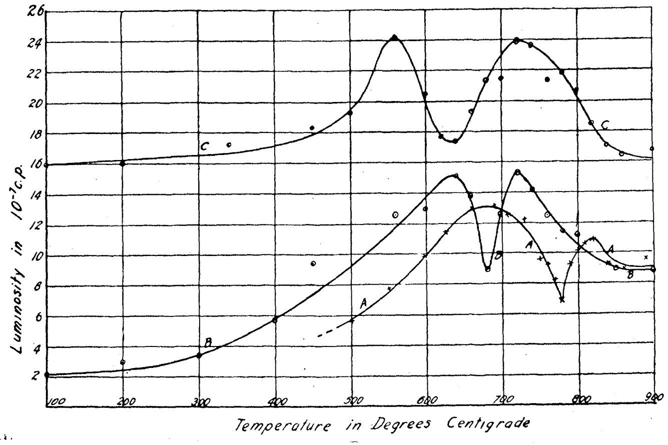

Fig. 4. Initial luminosity plotted against the temperature of the heat-treatment previous to exposure to the radium rays. Capsule No. 4 used in every case. Curve *A*, a sample of Kahlbaum; *B*, Mallinckrodt; *C*, Merck was used.

using the same source of radiation, capsule No. 4, and the three samples of barium bromide, Merck, Mallinckrodt and Kahlbaum. The number of inflection points and the general shape of the curves are the same in all cases, although the maxima and the minima are reached at different temperatures, and the brightness varies somewhat from sample to sample. It is proposed to continue the study of these variations in the luminescence with previous heat-treatment.

There is no point of inflection nor discontinuity in the curves near the temperature at which the water of crystallization disappears, nor does there seem to be such point at the fusion temperature, 880°C. The duration of the heat-treatment seems to have no effect. After a few minutes exposure at a given temperature, no difference can be found between the resultant initial brightness and that after the sample has been heated at this same temperature for days.

The barium bromide which has been exposed to the radiations appears to recover its initial properties on heating to 700°C or more. There is no observable difference in the variations of luminosity with temperature between samples which have been exposed to α-, β- and γ-rays for weeks and samples which have not been so exposed, provided they have been adequately heated.

If luminescence is explained by the destruction and the recovery of the active centers, this experiment shows that the α-, β- and γ-rays must have something to do with the production of these active centers as is indicated by the initial rise of luminescence as well as the destruction of the centers resulting in the decay which is always observed. These studies seem to add weight to Perrin's[5] assumption, that the luminescence must be affected by the bath of radiation in which the luminescing material is placed. He assumed that the radiation must also affect the recovery process. Witmer[6] using Perrin's hypothesis derived an equation to explain the facts found in Rodman's paper on the luminescence of pure radium bromide. Witmer's equation in its present form does not seem to fit the results of this study on pure barium bromide. To make this theory satisfy luminescence in general some revision must be made in it.

The writer wishes to acknowledge his indebtedness to Professor D. H. Kabakjian for his suggestions and his help in this studv.

Randal Morgan Laboratory of Physics,
University of Pennsylvania,
June 10, 1926.

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[5] J. Perrin, Ann. de Physique **11,** 5 (1919).
[6] E. E. Witmer, Phys. Rev. **24,** 639 (1924).