ACTION OF RADIUM SALTS ON GLASS.

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In 1908 the author began some experiments on the action of radium salts on glass, by sealing up a small amount of the salt in thick-walled glass tubes. These have been recently opened and examined, and in view of the interest now being taken in the formation of pleochroic halos in rocks, the results observed may be of some value.

The method employed was as follows:—

Some pieces of barometer tubing were selected, and a number of pieces about 5 cm. in length cut off. In some cases the radium salt was hermetically sealed in by fusing the ends of the glass, and in others the ends of the tube were merely plugged by paraffined cork.

The radium salt was one purchased from Messrs. Harrington in 1903, when it was stated to contain $\frac{1}{1000}$ of its weight of radium bromide. On being tested against black oxide of uranium, it has at the present time an activity about 5,000 times as great as that substance.

Two kinds of tubing were employed, one which showed a *very faint* pink tinge when looked at through the tube endwise, while the other had a pale greenish colour, but both colourations were very slight. After sealing the tubes were put away in a box, to which no light was admitted. Two control tubes of each sample were also used, one being kept in the dark and the other exposed to the brilliant sunlight of South Africa. This last was done because ordinary white glass exposed on the veldt acquires a dark amethyst tint in a few months. Unfortunately these samples were swept away in a storm.

On examining the tubes containing the radium from time to time, a progressive colouration was seen to be developed, more particularly at the end of the tube where the radium was lying, but it spread for some distance above. In one case, for example, the radium salt occupied a portion of tube to the extent of not more than 2 mm., while the colouration extended for 8 mm., diminishing gradually in intensity. On observing this the

tubes were occasionally turned over, so as to spread the colouration over as large a surface as possible.

FIG. 1.
M = 1.1 Diameters.

On making a close examination of these tubes a marked difference was seen in the manner in which the colouration had developed in the two kinds of glass; the photographs show this difference very clearly.

FIG. 2.
M = 1.5 Diameters.

Pieces of the radium-coloured tube and of the control sample were cut off, the ends ground and roughly polished, cover glasses being placed over

the ends and kept in place by a little cedar oil. The two tubes were mounted side by side in a cork and photographed together, a Zeiss micro-planar lens being used for the purpose.

Fig. 1 shows the two pieces of pink glass, and Fig. 2 the green. The dark patches in the left-hand tube in Fig. 2 are due to refraction at cracks in the glass. In Fig. 1 it can be seen that a dark zone is formed close to the bore of the tube, while in Fig. 2 the darkening extends to a greater distance without being so intense in any place. The real intensity of the colouration, as seen visually, is not correctly indicated by the photographs. The magnifications are about 1.1 for the pink tube and 1.5 for the green tube.

FIG. 3.

M = 3.5 Diameters.

On submitting No. 1 to a magnification of about 12 diameters a very remarkable appearance presented itself. The colouration was seen to have proceeded in a series of zones, one zone being clearly marked off from the rest, as shown in Fig. 3. Considerable difficulty was experienced in getting these zones clearly defined in the photograph, but this was accomplished at last by aid of isochromatic plates in conjunction with a light filter.

The innermost zone is very dark, while the succeeding zones show a gradual fading of the colour, which persists, however, right to the outside of the tube. The external diameter of the pink tube was 9.32 mm., the internal diameter 4.35 mm., and the thickness of wall 2.48 mm. The thickness of the first dark zone was 0.27 mm., and of the second 0.33 mm.

The zonal structure was not developed by the action of the radium, as it can be traced in the control tube, but not of course very distinctly, as the glass was not perfectly homogeneous, a sort of schistose structure being present. There was evidently some obstacle to the free passage of the α -particles, encountered at the junction of the layers of material.

The colouration extends into the glass for over 2 mm., and at all points of optical discontinuity a sharp change in the colouring power occurred, as though there was a strongly resistant layer at the junction of the two zones. From Professor Rutherford's paper (*Phil. Mag.*, Jan., 1910) one might conclude that the maximum depth of the colouration should be proportional to the range of the α -particles, and he gives the number 0.041 as the distance to which the α -particles should penetrate in glass.

If the α -particles fired into the innermost layer of the tube remain embedded there, could they impart a radio-activity to the layer, which in its turn would send off more particles, thus making the second layer active, and so on? This point might have been elucidated had the tube been examined regularly during the two years. An experiment has been started in which the tube will be examined at intervals.

The purple colouration I take to be due to some manganese compound, formed by the action of the rays on the glass in the one case, and in the other the increase in depth of the green colouration, to the possible reduction of a ferric compound to a ferrous one. Manganese in small quantities is frequently added to glass used for making tumblers, etc., in order to counteract the green colour due to iron compounds; the intense colouring power of the higher oxides of manganese is well known.

Plugging the tube with cork seemed quite as efficacious as hermetically sealing; the colouring did not proceed much beyond the cork. The photograph, Fig. 3, is of a corked tube, and the intensity of the colouration should be relatively greater than that shown, as the coloured portion of the tube was only 1.5 cm. in length, whilst the control tube was 3.2 cm.

The amount of emanation present in the tube cannot have been very large at any time, as, assuming the purity of the radium salt to be $\frac{1}{1000}$, this would give off a total amount of emanation which, in two years, would have approximately amounted to $\frac{1}{1000} \times .3 \times .1 \times .730$ cb. mm. of helium.* As this was continually decaying, and, also, in the case of the corked tubes, escaping, the amount present at any one time must have been very small, although it is most probable that the action is a cumulative one. The nature of the colouration clearly depends on the

* This would be the amount of emanation from a gramme of the salt. The actual amount used was only a few centigrammes.

nature of the glass. Further experiments are being proceeded with on various kinds of glass and other materials.

Professor Joly, in the *Phil. Mag.* for Feb., 1910, gives an account of an appearance similar to that just described, in a specimen of greisen. From the description of the sets of halos, it seems that a structure must exist in the crystal section similar to that occurring in the glass tube of my observations, but here again the action is limited to a small distance. A piece of glass tubing, owing to the method of manufacture, will probably be in a state of strain between the two surfaces, from which relief may be obtained by the zonal structure being formed. In the case of the crystal

FIG. 4.

M = 12 Diameters.

referred to by Professor Joly, since it separated from the magma, it would have been subject to some severe strain when the whole mass had become solid, on which a zonal structure may have been developed. The two surfaces of adjacent zones would naturally form boundaries to materials which are in some slightly different physical state, and so the α or other particles might experience a difficulty in crossing the boundaries. If the surface layers of the different zones were in a state of great tension they might act as though their densities were much greater than that of the mass of the material enclosed between their surfaces, and thus the α particles would experience a check to their velocity on crossing the boundary.

The foregoing was written in August, 1910, and since then observations have been taken upon other tubes in which impure radium salt had been sealed. The glasses used were: (1) a greenish coloured barometer tubing, (2) white thermometer tubing, (3) Jena combustion tubing, (4) tubing of the same kind used in the experiments detailed, (5) ordinary soft glass tubing. After introduction of the radium preparation the tubes were plugged with cork, and small cover-glasses fastened on the ends with Canada balsam. Control samples of tubing were cut off at the same time, which were kept in the dark. These were made of various lengths, so that any increase in the depth of colouring produced by the action of the radium could be contrasted with the original depth of colour by comparing the experimental tube with others of different length in the manner usual in colourimetric methods.

1. The length of the experimental tube was 3·3 cm., small plugs of cork 0·3 cm. closed each end, and the radium salt occupied about 0·3 cm. The control tubes were 4, 5, 6, and 8 cm., and the time taken for the depth of colouration to reach that shown by a greater length of control tube, which had not been acted upon by radium, found. This time cannot be stated with any degree of certainty, but a progressive colouration was shown, and the contrast presented the same difficulty as that found in comparing intensities of illumination of lights of different colour. The green colour was changed to a browner shade, especially close to the inner wall of the tube.

The following figures must be taken as approximate only:—

Time in months.	Length of control tube to match colour.
0	3·3
3	4·0
6	5·0
8	6·2
12	8·0

A brown layer was formed close to the inner wall of the tube, and this gradually merged into the general green colour of the glass and reached the outer wall. The thickness of the wall was 0·23 cm.

2. Jena hard glass tubing. This glass had a bluish green-colour, and no change occurred in twelve months in a tube 2·1 cm. in length.

3. White thermometer tubing 2·2 cm. in length. This specimen was nearly white. A faint violet colouration was seen in the layer immediately surrounding the bore, and a darkening occurred throughout the mass. The length at the end of twelve months of the control tubing which matched that of the experimental tube was 5 cm.

4. This specimen was of the same kind of glass as that referred to in the first part of the paper. The tube was 2 cm. in length, and the free space when plugged about 1.5 cm. The radium salt got mixed up with the balsam used for sealing, and was rather irregularly spread over about 0.3 cm. The action extended along the tube for a greater distance than that occupied by the radium, even penetrating to the portion covered by the cork. With this tube the progressive colouration could be followed and the time for each successive zone to be developed noted.

In 20 days a decided colouration was visible, and this extended for a certain distance and then stopped abruptly at the end of the first layer of the glass. In 53 days the second zone could be traced and was quite distinct at the end of 120 days, when the third zone had begun to show itself. In 250 days the third zone was well marked, and at the present time, 337 days from the commencement, five zones can be seen. The colouration does not yet extend right through the wall of the tube. The width of the zones does not change with time, but the depth of colouration does.

The measurements made on the tube were as follows:—

Internal diameter	3	mm.
External „	8.5	„
Thickness of wall	2.5	„
1st and deepest coloured zone	0.05	„
2nd zone	0.1	„
3rd „	0.3	„
4th „	0.5	„

The width of a zone is not uniform, as it depends upon the original zonal structure present in the glass, and the values given are average ones, as a reference to Fig. 3 will show. The zonal structure is not developed by the radio-active action.

5. This was a specimen of fairly homogeneous glass, without zonal structure, and it showed a progressive depth of colouration from the inner to the outer wall. The depth of colour developed in a given time was greater than that of any other glass examined.

From the papers of Professors Joly and Rutherford it might be inferred that they consider the colouration as being due to the α particles alone. If this is so, then the range of the α particles must be much greater than that indicated, for there is no question as to the depth of penetration in the glass tubes. But why is the development of the colour to be limited to the α particles? The β and γ rays pass through the material and clearly must exert some influence; in fact, the total change must be due to this combined effect.

It seems a matter of interest to know that very small amounts of a radium salt can give rise to such definite changes in colour. A tiny speck of the radium salt, weighing less than a milligram and very impure, will show a distinct colouration in a capillary tube at the end of 40 days. The small amounts of radium present in ordinary rock may thus well be capable in geological time of giving rise to the pleochroic halos.

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