S Y N O P S I S

INTRODUCTION

The discovery of radioactivity has brought about a revolution in our ideas in physics, and also in our ideas about the energy content of the earth. The earth has at its disposal a continuous energy source in its interior which originates from the disintegrating nuclei of the radioactive constituents of the earth's crust. In addition to the heating produced by this radiation, which controls the heat content of the earth, we know from experiments that other effects are produced in solids, such as ionization, colouration, luminescence, thermoluminescence, and even the destruction of the crystal structure of the solid. The extent of these effects one can find in Nature depends on the mineral investigated and on the intensity of the radiation itself.

It was Pearsall (1830) who first showed that a colour was produced by the action of an electric spark on a colourless fluorite crystal, and this corresponded to the natural colour of fluorite; thus a possible explanation of the pale colours of minerals was discovered. He coloured a fluorite crystal, rendered colourless by heating, blue to pink with the spark from a Leyden jar. According to him "Could it not be that the natural fluorite owes its colour to a special structure Could one not suppose that Nature employs the same means and that it is electricity which produces the colour in the natural state Both the natural and artificial colour are destroyed by heat, and the colour as well as the phosporescence can be restored repeatedly by electricity".

One can not doubt that in his experiment it was the short wavelength ultraviolet light from the spark that produced the colour and the phosphorescence, which was in fact the thermoluminescence. If one substitutes the word radiation for electricity in the expression, one can see that Pearsall was as near to the present day explanation of colouration and thermoluminescence as might be expected before the discovery of radioactivity. The colouration of certain minerals is particularly indicative of a radioactive effect, and the first suggestion that the natural colouration of minerals is caused by radioactivity was due to Siedentopf (1905). The effect of natural radioactivity on the luminescent properties of minerals was first recognised in thermoluminescent minerals, i.e. those which glow on heating, and it was assumed that the initial excitation energy was supplied by the rays from the radioactive substances.

The study of colouration and luminescence has given an insight about the structure of solids, and of the mechanism of light emission. For the mineralogist and an earth scientist, the natural colours of minerals produced by radiation, as well as their colourability under artificial conditions, can be of importance in many respects. The former are the results of a physical process which has been operative throughout geologic time, and therefore give, with correct interpretation, information about those processes in the strata; the later can show up variations which could not be detected otherwise. Luminescence analysis is very suitable for the identification of traces of impurity which can influence the structure of the mineral, and can, under certain conditions, depend on the age of mineral and its mode of formation. From experiments on the luminescence and thermoluminescence of fluorite, an explanation has been given of the dependence of the rare earth content on the mode of formation of the fluorite and their distribution in this mineral (Przibram, 1956).

The results in this field are not so simple to interpret, as it is not only a question of the presence in the mineral of an element which luminesces or the mineral produces thermoluminescence, and of how it is built into the crystal, but also of the thermal pretreatment of the specimen, the presence of impurities, and the effect of radioactivity. The present studies form a part of such investigations which would in a small way reveal the mystery of the effects of radioactive radiations on crystals.

PRESENT STUDIES

Natural fluorite exhibits variety of colours, and is a sensitive and reproducible thermoluminescent mineral. The present work deals with the studies carried out on naturally occurring fluorites, exhibiting five different colours viz., light green, blue, violet, yellow, colourlees, limestone and quartz, collected from Amba Dungar Mine, in Gujarat, India and pale green fluorite and limestone from Belamu hill, in West Bengal, India. The studies covered in the present investigation deal with various parameters, viz., the effects of heat, irradiation, crystallization temperatures etc. on thethermoluminescent characteristics of the samples and their emission and absorption spectra. In addition to these a new method based on computer aided graphical curve matching method has been suggested to determine the glow curve parameters, viz., activation energy, frequency factor and half life.

A simple technique has also been suggested to generate 3-D isometric glow curves. Besides these the role of thermoluminescence of the samples as an aid to geochronology has been investigated.

EXPERIMENTAL METHODS

Thermoluminescence of the samples was recorded with the experimental set-up similar to that described by Kaul et al. (1966). To avoid the variation in TL-glow due to grain size, samples with grain size 88-149 µm were thoroughly mixed and uniformly spread on the TL-oven. The thermal background was reduced by introducing a filter between the sample and the photomultiplier. The heating rate used during the TL-glow recording was generally 10°C/sec. In course of some investigations this was changed according to the experimental requirement. The radioactive sources used for irradiating the said samples were a Co^{60} source for gamma irradiation (dose rate 1330 rads/minute) and an Am^{241} source for alpha irradiation. The gross alpha activities of the samples were determined by the 'Thick source counting technique' as adopted by Turner et al. (1958), using ZnS as detector on a disc of 54mm diameter. The absorption spectra of the fluorites were recorded with the help of a Beckmann (Model-26) double beam absorption spectrophotometer, while the emission spectra were made by monochromator (Model 82-410).

RESULTS AND CONCLUSIONS

From the present studies it is found :

1) The Tl sensitivity for different coloured fluorites, collected from Amba Dungar is different. This is attributed to variation in the impurity concentration in each sample.

2) Glow peak temperatures for different coloured samples do not show any significant change, indicating thereby that the peaks are due to host lattice and not due to the impurities present, even though the impurities play an important role in their relative TL - intensity.

3) Characteristics of TL glowicurves of Amba Dungar fluorites bear resemblence to the ones from a hydrothermal deposit, while the characteristics of Belamu fluorite are akin to the ones from pegmatites.

4) TL glow peaks of fluorites are a combination of two or even more peaks and not one single peak as observed in the ordinary two dimensional record.

5) The TL characteristics of the samples have been found to change after annealing, while the effect of such treatment is not same for all the peaks. It is concluded that annealing produces more defects or trapping centres, which are responsible for increasing the TL intensity, it also destroys some of the trapping centres, which cause the reduction in TL intensity of some glow peaks, and also, some trapping centres remain unaffected as no change is found in the TL intensity of the glow peaks attributed to such centres.

6) The studies on the TL kinetics and trapping parameters of the fluorites indicate that the TL peaks in fluorite follow the second order kinetics.

7) A programme is developed to determine the peak parameters viz., activation energy, frequency factor, and half life time, while attempting computer aided graphical curve matching method.

8) From the present studies on the absorption and emission spectra of fluorites, it is concluded that blue colour owes its origin to Y^{3+} ion associated with F^- centre, yellow to O_3^- molecular ion, green to Sm^{2+} ion and violet to metallic calcium particles. These findings are in confirmity with the findings of earlier workers.

9) From the glow curves recorded through various filters a 3-D isometric glow curve is generated by a computer programme. The 3-D isometric glow curve of the green fluorite is the most complex amongst the four coloured varieties while the yellow variety shows the least

complexity. As the number of impurities shown by the green variety of fluorite is more than the yellow variety, it may be concluded that the complexity of 3-D glow curves is directly related to the number of impurities acting as emission centres.

10) Homogenisation temperatures of fluorites from two contrasting geological environments are investigated by fluid inclusion thermometry as well as measuring the (Tu/Th) ratios from their characteristic thermoluminescence glow curves. Temperature of formation of fluorites thus obtained confirm the findings made by geological studies conducted in these areas by various workers.

11) The TL age estimates made in course of present studies are very low compared to the geological ages of the samples as reported by various workers. TL ages of different coloured fluorites collected from Amba Dungar do not vary much from one coloured sample to another. This may imply that all the varieties of fluorites crystallised practically at the same time. TL age of Belamu fluorite is much less than the age of Amba Dungar fluorites. Even though the TL age estimates work out to be less than geological estimates, the sequence of formation of fluorites, quartz and limestones agree well with the geological findings. The lower estimates of the TL ages are attributed to the dependence of TL on too many factors viz., radioactive content of the material, availability of trapping sites, temperature and pressure etc. Thus the age estimate made in the present investigation can represent only the last phase of the geological processes.