Effect of deformation on the thermoluminescence of BaSO$_4$:Tb phosphors

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Received 30 December 2010; revised 17 February 2011; accepted 31 March 2011

Thermoluminescence (TL) of BaSO$_4$ phosphors has been studied. BaSO$_4$:Tb phosphors having different concentration of Tb were prepared by solid state reaction method and their TL has been studied after gamma irradiation. Two distinct peaks are observed in the TL glow curve, one around 150°C and another around 210°C. It is also observed that TL peak at 150°C increases with gamma dose given to the samples. However, TL peak around 210°C saturates for gamma dose higher than 0.930 kGy. In order to study the effect of deformation on the TL, a load was dropped on to the sample with different impact velocities. TL intensity of both the peaks decreases when the TL glow curve is recorded after deforming the irradiated samples. TL emission spectrum and photoluminescence (PL) study on the sample show two prominent peaks around 480 and 550 nm due to $^5D_4 \rightarrow ^7F_6$ and $^5D_4 \rightarrow ^7F_5$ transitions of Tb$^{3+}$ ions. It is suggested that the recombination of electrons released from the traps during the thermal excitation is responsible for the luminescence in this system and the deformation of sample damages the traps centers which reduces the TL intensity of the sample.

Keywords: Thermoluminescence, BaSO$_4$

1 Introduction

Thermoluminescence (TL) is the emission of light from a pre-irradiation material on stimulating thermally i.e. by heating which induces relaxation of electronic charges resulting in radiative recombination causing luminescence. The luminescence is at cost of energy absorbed from ionizing radiation where the charge carriers (electron and holes) are produced by ionization get trapped at the sites caused by imperfection\(^1\).

Thermoluminescence of BaSO$_4$ phosphors has been studied by many researchers\(^2\)\(^-\)\(^5\). The temperature of TL glow peaks induced by a rare earth ion in BaSO$_4$ lattice is found to be determined only by the valance state of rare earth\(^6\). It was proposed that radicals such as SO$_4$$^-$$^-$, SO$_3$$^-$$^-$, O$_3$$^-$$^-$ and so on produced by irradiation from stable trapping sites of varying activation energy that are responsible for the complex multi-peak thermoluminescence (TL) glow curve\(^7\).

Recently, Laxmanan\(^8\) reported the effect of pressure on the luminescence properties of gypsum, anhydrite, calcite and Dy doped CaSO$_4$ sample. It is concluded that the changes observed in TL should be attributed to the damage of traps/luminescence centers rather than to the damage of their crystal structure.

The present paper reports TL, photoluminescence (PL) and the effect of post irradiation deformation on the thermoluminescence of BaSO$_4$:Tb phosphors.

2 Materials and Methods

The BaSO$_4$:Tb samples have been prepared by solid-state reaction method, in which the requisite amount of AR grade BaSO$_4$ and Tb$_2$O$_7$ (as per mole concentration) were mixed and heated up at 800°C in porcelain crucible in open air for 24 h, then cooled very slowly. It was then annealed at 750°C for one hour and quenched to room temperature. XRD pattern of the prepared sample is recorded and it confirms the formation of the crystal. Samples were exposed to gamma rays using $^{60}$Co source having the exposure rate of 0.93 kGyh$^{-1}$. TL glow curves were recorded by usual set-up consisting of a small kanthal strip for heating the samples, temperature programmer, photomultiplier tube (931B), dc amplifier and an X-Y recorder. An aliquot of 1 mg of phosphor was heated every time at a heating rate of 90°C min$^{-1}$. Samples were deformed by dropping a load of mass 0.4 kg from different heights onto it. The apparatus used for TL spectral study consist of a continuous scanning tap of 0.25 m focal length Jerrel Ash monochromator, with an aperture (speed) of F/3.6 and linear dispersion of 3.3 mm per nm. The photoluminescence (PL) emission spectra of the samples were recorded by using fluorescence spectrophotometer (Shimadzu RF-530 XPC). Emission was recorded using a spectral slit width of 1.5 nm.
3 Results and Discussion

Figure 1 shows TL glow curves of BaSO₄:Tb phosphors having different concentration of Tb. Two distinct peaks are observed for all samples. Maximum TL is observed for 0.05 mol % doping of Tb ions. The undoped BaSO₄ shows very weak TL.

The number of active luminescent centers is expected to increase with concentration of Tb. As a result, glow intensity increases with concentration. However, the glow intensity cannot be expected to increase indefinitely with concentration, since the rate of formation of active luminescent centers by capturing the holes during the irradiation, might be fading rapidly and concentration is affected. It is also observed that the temperature corresponding to main TL peak in the TL glow curve of BaSO₄:Tb phosphors slightly shifts towards low temperature side with increasing concentration of Tb. This behaviour can be explained by assuming that either a multilevel or a continuous distribution of trap depth is associated.

Figure 2 shows the dependence of TL intensity of BaSO₄:Tb(0.05 mol %) phosphors on gamma dose given to the sample. Peak intensity increases with increasing gamma dose, both peaks shift towards lower temperature side with increasing gamma dose. TL intensity of 150°C peak increases with gamma dose in the range studied, however, the TL intensity of 210°C peak seems to saturate at higher gamma dose. Generally, at higher gamma dose given to the sample the peaks at higher temperature arise. In present paper, it is found the peak at low temperature increases with increasing gamma doses given to the sample and the peak at higher temperature has become saturated for higher gamma dose. Similar results have also been reported for thermoluminescence in X-ray irradiated Mn doped BaSO₄ phosphors. The increase in TL intensity with respect to gamma dose is attributed to the increase of active luminescent centers with gamma irradiation. The saturation of TL can be explained on the assumption that only limited numbers of trivalent Tb ions are available for charge reduction with increasing gamma dose. Gamma dose response curve of 150 and 210°C TL peak is given in Fig. 3. TL peak at 150°C increases almost linearly in the range of gamma doses (0-1.86 kGy) given to the sample.

Figure 4 shows the effect of post irradiation deformation on the TL of BaSO₄:Tb(0.05 mol %) phosphors. No drastic change in glow curve structure was seen. However, the intensity of both the peaks decreases with increasing impact velocity of the piston used to deform the sample. The decrease in 210°C TL peak is more as compared to 150°C peak. The deformation subsequent to irradiation may modify the existing traps, creates new but unfilled traps and/or cause a redistribution of electron or holes along there traps due to passage of recombination in the vicinity of traps. In the present study, it seems that the de-trapping of trapping centers may be responsible for decrease in TL intensity with deformation of the sample.
Figure 5 shows the TL emission spectra of Tb-doped BaSO\(_4\) phosphors of un-deformed and deformed samples. It is found that the emission wavelength is the same in all the curves, however, TL peak intensity decreases as the height of the load dropped onto the sample (to deform it) is increased.

In order to know the behaviour of the luminescence centers in BaSO\(_4\):Tb phosphors, the photoluminescence of BaSO\(_4\):Tb (0.05 mol %) samples (Fig. 6) has been recorded. The emission spectrum after excitation by 385 nm shows two prominent peaks at 478 nm and 594 nm which are characteristics of Tb\(^{3+}\) ions due to \(^5\)D\(_4\) → \(^7\)F\(_6\) and \(^5\)D\(_4\) → \(^7\)F\(_5\) transitions of Tb\(^{3+}\) ions.

For TL mechanism alkali earth sulphate, Luthra et al.\(^9\) proposed that trapping and recombination are the characteristics of host lattice. When BaSO\(_4\):Tb phosphors are exposed to gamma rays, the trivalent dysprosium acting as electron traps gets induced to divalent terbium with the production of trapped holes centers (like SO\(_4\)^2\(^-\) and SO\(_2\)^2\(^-\)). The holes are released from thermal excitation and recombination of holes with electron at Tb\(^{2+}\) sites. Luminescence is observed during de-excitation of excited Tb\(^{3+}\) ions.

4 Conclusions

In BaSO\(_4\):Tb phosphor, the TL peak around 150°C may be more useful for dosimetric purpose because of almost linear dose response and less mechanical bleaching effect.

References