Thermoluminescence Studies of Gamma Irradiated Sodium Chloride Single Crystals and Microcrystalline Powder Doped with Terbium

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ABSTRACT

This paper reports the thermoluminescence of $\gamma$ - irradiated terbium doped NaCl single crystals and powder. The NaCl crystals having different concentrations of terbium were prepared by melt technique (slow cooling). The crystals of small sizes were cleaved from grown crystal block and crushed to obtain powder or microcrystalline powder. The annealed samples were irradiated by gamma source with dose rate of 0.50kGy/hrs. In thermoluminescence of NaCl:Tb crystals and powder samples only one peak is observed in the glow curve at around 210-245\textdegree C.

Keywords: Thermoluminescence, $\gamma$–irradiation.

INTRODUCTION

When a crystal is warmed at a constant rate in the dark after being exposed to excitation at low temperature it is found to exhibit luminescence. This type of luminescence is called Thermoluminescence (TL). Thermoluminescence of single crystals and microcrystalline powder of alkali halides have been studied by various research groups (Ausin 1972, Patley 1989, Sastry 1985). Davidson et al. (2002, 2004,
2006) measured the thermoluminescence of a number of NaCl and KCl crystals following irradiation at ambient temperature with the same dose (10kGy) of Co-60 rays. They compared the TL of pure samples and the samples doped with europium and calcium ions.\textsuperscript{1-6} The behaviour of single crystals of NaCl: Ca\textsuperscript{2+}, Mn\textsuperscript{2+} exposed to gamma rays was studied by A Ortiz \textit{et al.}\textsuperscript{7}. Recently Bangaru and Muralidharan (2009, 2010, 2011&2012) and P.M. Bhujbal \textit{et al.} (2012) also reported the enhanced luminescent properties and thermoluminescence studies in alkali halides by doping rare earth materials\textsuperscript{8-14}. Studies on microcrystalline powders could be interesting as the powders can be thought of made up of two physically intertwined subsystems; one the perfect lattice preserved owing to cleavage and the other the imperfections which bind small indestructible regions within each crystallite. TL studies of microcrystalline powders and its comparison with the results of single crystals give useful information about the interaction between colour centres and other grosser imperfections such as grain boundaries, dislocations and surface defects. Mohril \textit{et al.} (1977), Wakde \textit{et al.} (1979), Deshmukh \textit{et al} (1984, 1986) reported several interesting differences in the results on colour centres in microcrystalline powders from the single crystals\textsuperscript{15-18}. We have reported mechanoluminescence (ML) of gamma irradiated single crystals and microcrystalline powder of terbium doped NaCl in our previous studies\textsuperscript{19}. The present paper reports TL of gamma irradiated Tb doped NaCl single crystals and microcrystalline powder.

**EXPERIMENTAL**

Single crystals of NaCl doped with different concentrations of Tb were grown by melt method. Analar grade chemicals were used in the present investigation. The crystals of small sizes (1x1x1, 2x1x1, 2x2x1, 1x2x3mm\textsuperscript{3} etc) were cleaved from the grown crystal blocks. Microcrystalline powders were prepared by crushing some of the grown crystal blocks. Test sieves of known dimensions were used to separate the microcrystalline powders of different grain sizes. The crystals so grown and the microcrystalline powder as obtained were annealed at 450°C for two and half hours and subsequently cooled to room temperature. The irradiation of samples was carried out using \textsuperscript{60}Co gamma source having exposure rate of 0.50 kGy/ hour. Thermoluminescence measurements were carried out of undoped and Tb doped NaCl single crystals and powder samples using a PC based TLD reader (Nucleonix Model 10091). The TL measurements were taken after 24 hours of irradiation and for the measurement of TL intensity of single crystals of known mass of different sizes (reading normalized to same mass) and microcrystalline powder of mass 2 mg of gamma irradiated phosphor was heated every time at a heating rate of 5°C/sec. For each measurement at least three observations were taken.

**RESULTS AND DISCUSSION**

A single glow peak for undoped and doped samples are obtained at around 210-245°C for single crystals and powder samples at heating rate of 5°C/sec (Figure1&2). The TL Intensity of single
crystal is less than that of the microcrystalline powders of the same mass. The microcrystalline powders of small size samples have lower intensity than the large size samples. Figure 3 shows the thermoluminescence glow curve of undoped and Tb doped NaCl microcrystalline powder (0.120-0.150μm) of different concentrations (Dose: 0.50kGy). It has emerged in the last thirty years that many of the thermoluminescence process observed in alkali halides irradiated at any temperature (glow curves) are probably the result of the thermal release of holes from different traps, which recombined at the F-centers or the recombination of radiation induced lattice defects such as interstitial halogen atoms and vacancy centers (Alvarez-Rivas 1980) or both. The less intensity in the small size powder may be due to the fact that small grain of the NaCl: Tb cannot hold interstitial clusters as much as in large grains of the sample.

Figure 4 shows the Variation of total TL intensity of γ -irradiated NaCl crystals with different concentrations of Tb (Dose: 0.50kGy). It is observed that TL intensity initially increases with increasing the concentration of dopant attains an optimum value for a particular concentration (200ppm) and then decreases with further increase in the dopant concentration. The effect of incorporating rare earth impurities into insulating host crystals during crystal growth involves several considerations which include whether interstitial or substitutional sites are occupied, the clustering of impurities, possible precipitation of impurities to form new phases and the question of charge compensation of the impurities ions. In the present work doping of Tb$^{3+}$ lead to charge compensating vacancies in NaCl, which increases the probability of formation of colour centers. The trivalent impurity ion Tb$^{3+}$ enters the crystal replacing Na$^+$ ions. During γ – irradiation electron hole pairs are created as expected hole is captured by host related centers and some of the released electrons are captured by the impurity Tb$^{3+}$ ions reducing these to Tb$^{2+}$ ions in addition to the formation of F-centers. So γ -irradiated Tb doped NaCl crystals, Tb ions enhance the relative density of defects. Therefore the TL intensity initially increases with the increase in the concentration of rare earth ions as more luminescence centers are created and attains an optimum value for a particular concentration (200ppm) and then decreases with further increase in the dopant concentration. However, the TL intensity cannot be expected to increase indefinitely with concentration, since the rate of formation of active luminescent centers by capturing the holes during irradiation might be fading rapidly and concentration quenching is occurred.

Fig 1: TL glow curve of NaCl: Tb single crystals for different concentration of dopant gamma dose 0.50 kGy.
Figure 5 represents the dependence of TL intensities of NaCl: Tb crystals on various doses. The TL intensity of γ-irradiated Tb doped NaCl crystal and microcrystalline powder initially increases with γ-dose given to the samples and seems to saturate at higher gamma doses (1.0kGy). The increase in TL intensity with increasing γ-dose may be due to increase in number of active luminescent centers with γ-irradiation and subsequent emission of TL due conversion of Tb$^{2+}$ ↔ Tb$^{3+}$ during heating. The dose saturation of TL can be explained on the assumption that only limited number of Tb ions is available for charge reduction with increasing γ-irradiation.

The TL intensities of microcrystalline powders are more than for crystals. The reasons for the observed differences in the intensities for crystals and microcrystalline powders might result from the additional number of trapping sites available in the powders or the greater defect mobilities due to surface /and or dislocations.
CONCLUSIONS

We have investigated the TL of NaCl doped with terbium. The TL results show variation in the peak temperature and total TL intensity with dopant concentration, particle size and gamma ray irradiation. The TL intensity obtained is more for microcrystalline powder than single crystal. The TL intensity of microcrystalline powder increases with particle size. TL emission in the present samples is induced by the gamma ray irradiation and depends on gamma ray doses, and maximum intensity is obtained for low concentration of impurity, which is a good characteristics for the development of materials for radiation dosimeter (cost wise) so phosphor NaCl:Tb may be use in thermoluminescence dosimeter.

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