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Mechanoluminescence studies of gamma irradiated potessium chloride single crystals and microcrystalline powder doped with terbium

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Abstract

This paper reports the mechanoluminescence (ML) induced by impulsive excitation of γ - irradiated terbium doped KCl single crystals and powder. The KCl 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. Mechanoluminescence is excited impulsively by dropping a load of 0.4 kg with impact velocity 313 cm/sec on to it. Two peaks are observed in the ML intensity versus time curves for both single crystals and powder samples of different sizes. In the ML spectra a broad band with single peak at 482 nm is obtained. The ML intensity of powder sample is less as compared to crystal sample of same mass. The ML intensity of microcrystalline powder increases with particle size.

Keywords: *Mechanoluminescence, Thermoluminescence, γ -irradiation*

1 Introduction

Radiation causes excitation of molecules besides formation of free radicals. The presence of excited states can be discovered by measuring of light emission when crystallite material is heated (thermoluminescence), or stimulated by light (photoluminescence), or when dissolved in suitable solvent(lyoluminescence). Mechanoluminescence is an interesting luminescence phenomenon whereby light emission in solids is caused by mechanical stimuli such as compressing, stretching, fracture, cutting, cleaving, breaking, grinding, rubbing, scratching, and crushing and so on [1–3]. The mechanoluminescence (ML) phenomenon has generated extensive research over the years because of its potential application for damage detection[4]. Recently Bangaru and Muralidharan (2009,2010,2011&2012) and P.M. Bhujbal et al.(2012) also reported the enhanced luminescent properties and thermoluminescence studies in alkali halides by doping rare earth materials[5-11].The present paper reports ML of gamma irradiated Tb doped KCl single crystals and microcrystalline powder.

2 Experimental

Single crystals of KCl 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 were cleaved from the grown crystal blocks. Microcrystalline powders were prepared by crushing some of the grown crystal blocks. Test sieves were used to separate the microcrystalline powders of different grain sizes. The crystals so grown and the microcrystalline powder of different sizes (75- 180μm) as obtained were annealed at 450^0C for two and half hours and subsequently cooled to room temperature.The irradiation of samples was carried out using ^{60}Co gamma source having exposure rate of 0.50 kGy/hour. The ML was excited impulsively by dropping a load of mass 0.4 kg on to the gamma-irradiated sample placed on the Lucite plate from the height of 50 cm using a guiding cylinder. The impact velocity of the load is determined by the relation $v = \sqrt{2gh}$. The ML was monitored by RCA 931 photomultiplier tube positioned below the Lucite plate and connected to storage oscilloscope (Scienific SM-340). ML emission was recorded by inserting filters of different wavelength between lucite plate and photomultiplier tube.

3. RESULTS AND DISCUSSIONS

Figures 1 shows time dependence of ML intensity of gamma irradiated KCl single crystals and powder doped with different concentration of Tb. Where, two peaks are observed in the ML intensity versus time curve [12]. The ML intensity of first and second peak as well as total ML intensity increases with increasing concentration of dopant up to 200ppm without any considerable change in t_m i.e. time corresponding to ML peaks. Thereafter the ML intensity decreases with further increase in concentration of dopant. Similar results are observed for KCl: Tb powder samples (Fig. 2); however the ML intensity of powder sample (120-150µm) is less as compared with crystal sample(of the same mass). In γ -irradiated rare earth doped alkali halide crystals, rare earth ions enhance the relative density of defects. That is why the ML intensity increases with dopant concentration. Initially the number of luminescence centers and defect centers increases with increasing concentration of dopant, which causes increase in ML intensity with increasing dopant concentration. For the higher values of dopant concentration, concentration quenching of luminescence centers may take place and therefore, the ML intensity decreases with further increasing in concentration of the dopant. As the matter of the fact, the ML intensity is optimum for a particular concentration 200ppm of Tb(dopant).

Figure 3 represents the dependence of ML intensities of KCl:Tb microcrystalline powder of various sizes. The ML intensity of microcrystalline powder decreases with decrease in the particle size.

The decrease in ML of microcrystalline powder than single crystal could arise for several reasons. In microcrystalline powder, large number of grosser defects such as dislocations, grain boundaries, etc., will be produced during crushing of single crystals. This will increase the surface to volume ratio of the system. During irradiation the colour centre production may get affected by the presence of these surface and dislocations (defects).This could be the reason for the less luminescence intensity in the small size microcrystalline powder. Lesser stability of the colour centres produced after irradiation in smaller size microcrystalline powder could be the another reason for less luminescence intensity. Further the dislocations (defects) may increase the diffusion which leads to increased recombinations which are inefficient to yield good ML intensity. One the other hand these dislocations may also act as sink for some defects responsible for ML and thus reduce the number of defects and hence the weak ML intensity. Further the adsorbed gases and moisture, on the surface may lead to non-radiative recombinations which in turn reduce the intensity yield in ML of microcrystalline powder. Moreover we may expect that these effects will go on increasing with the decreasing particle size and hence the less ML intensity with decreasing particle size is expected.

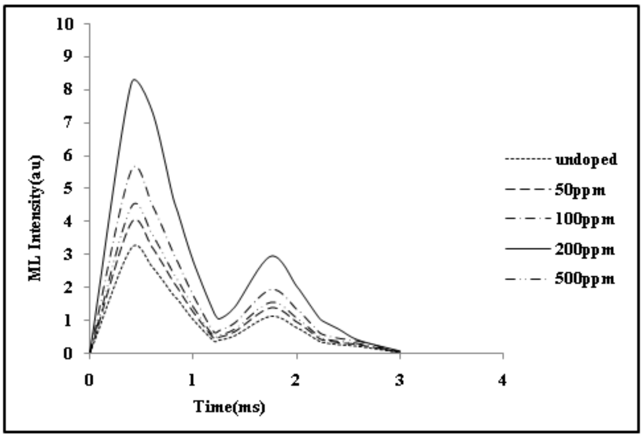
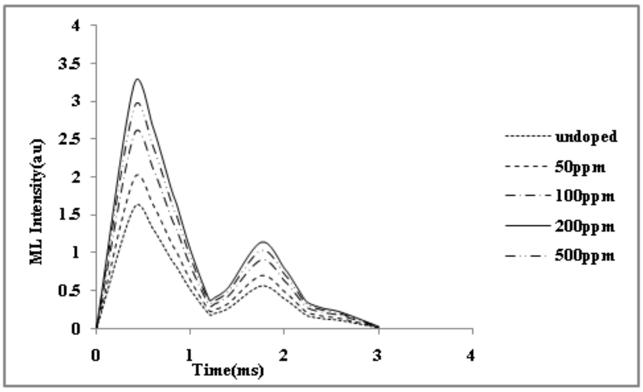


Fig 1: Time Dependence of ML Intensity of KCl:Tb Single crystals(0.25kGy)



.Fig 2: Time Dependence of ML Intensity of KCl:Tb powder(120-150µm)

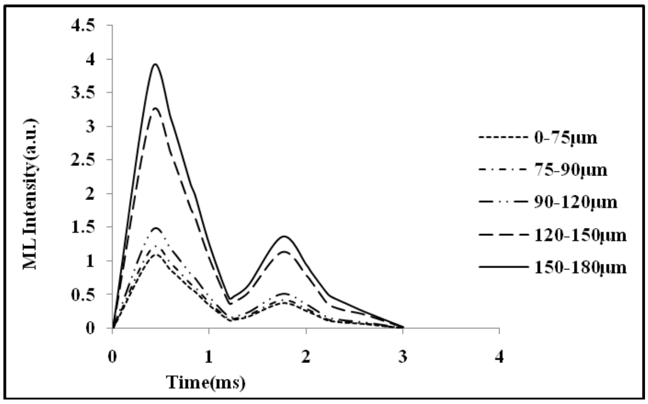


Fig 3: Mechanoluminescence glow curve of Tb doped KCl microcrystalline powder(200ppm) (0.25kGy)

Figure 4 represents the ML spectra of single crystals of KCl: Tb. The ML spectra consist of a broad band with single peak at 482 nm for doped crystals. The luminescence efficiency gets enhanced by the incorporation of Tb-ions in the KCl matrix. It has been reported that the spectrum of a given RE^{3+} ion is essentially the same in various host lattices. The major emission in Tb^{3+} doped is due to the transition $^5D_4 \rightarrow {}^7F_J$ which are mainly in the green region and often there is a considerable contribution to the emission from the higher- level emission $^5D_3 \rightarrow {}^7F_J$, mainly in the blue region.

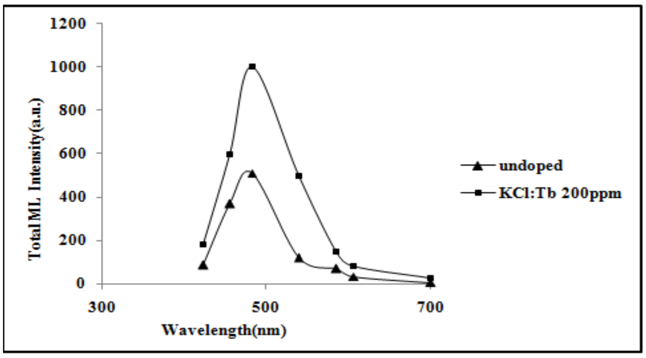


Fig 4 : ML Spectra of γ –irradiated KCl: Tb (200ppm)crystal.

In the present study we noticed that in the ML spectra of KCl :Tb, the effect of incorporation of terbium is the overall intensity enhancement in the host emission from 423nm to580nm takes place with broad peak at 482nm.It may either be due to the overlapping of emissions of host and impurity or due to the emission of host which is being enhanced by the impurity(Tb^{3+}). Figure 5 represents the Dose dependence of single crystals of KCl: Tb. It is noticed that both the total ML intensity initially increase with increasing irradiation dose, and then attain saturation values around 1000Gy. The increase in ML intensity may be due to increase in the density of defect centres. The saturation of the intensity can be explained on the assumption that only limited numbers of Tb^{3+} ions are available for charge reduction with increasing gamma dose.

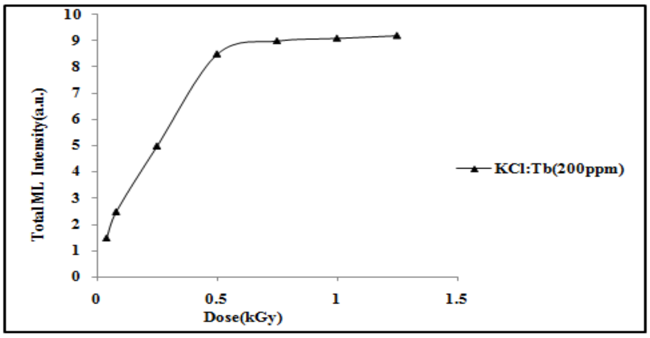


Fig 5: Dependence of total ML intensity of single crystals of KCl: Tb on different doses (Impact velocity 3.13 ms^{-1}, mass of piston 0.4 kg).

It is suggested that ML of KCl: Tb is strongly related to the movement of dislocations and the recombination of activated electrons and holes. The movement of dislocations excites carriers from the filled traps and the subsequent recombination of the electrons and holes in luminescence centers (Tb^{3+}).

4. Conclusions

We have investigated the ML of single crystals and microcrystalline powder of KCl doped with terbium and we found that two peaks in ML glow curve. The first peak lies in the deformation region and the second peak lies in the post deformation region. The Peak and total ML intensity is dependent on dopant concentration of terbium and gamma ray irradiation doses. the ML intensity of powder sample is less as compared with crystal sample(of the same mass).The ML intensity of microcrystalline powder increases with particle size.The ML spectra of doped crystals with broad peak at 482 nm signify the participation of terbium ions (impurity) in the ML phenomenon. ML emission in the present samples are 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 (costwise) so phosphor KCl:Tb may be useful in mechanoluminescence dosimeter.

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