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THERMOLUMINESCENCE BEHAVIOUR IN THE KBr : Sm^{3+} Tb^{3+} SINGLE CRYSTALS

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Abstract:-*KBr: $Sm^{3+}Tb^{3+}$ crystals were successfully grown by Bridgemann Stockbarger technique. Thermoluminescence (TL) properties of these crystals were investigated. Before taking the TL ,the sample annealed at 400° C and then irradiated at a γ –ray dose of 5 Gy. The TL glow curve was recorded after the samples were irradiated with γ-rays for 1-hour with the heating rate of 5°C/sec. The TL glow peak deconvoluted using the Origin 5.0 software. The deconvoluted glow peak reveals two clearly visible distinct peaks at temperature around 448K and 501K. The various parameters such as geometric factor, Activation energy and Frequency factor were found out using Chen's peak shape method. The calculated kinetic parameters show that both peaks follow the second order kinetics. The high temperature peak at 501K is due to high energy traps and it is very useful for TL dosimeter (TLD) phosphor characteristics. The deconvoluted TL emission spectrum for the glow peak at 501K shows two well separated peaks centered at 583nm and 644nm which confirms emission is only due to samarium ions. Scanning electron micrographs (SEM) of KBr: Sm^{3+} Tb^{3+} crystals, it is found the particles are cubic in shape with various sizes ranging from few microns to 10 microns and also cluster is found. The micron size of crystallite from SEM image is best suited for dosimetric applications.*

Key words*: KBr, Sm^{3+}, Tb^{3+}, TL, SEM*

1. Introduction

In recent years, thermoluminescence has been of interesting behaviour to study in single crystals. Thermoluminescence (TL) is being broadly used in examining the nature and behaviour of defect centers in solids, especially in ionic crystals such as doped and undoped alkali halides. TL process is the stimulated radiative recombination of released electrons, initially trapped by some defect centers in the material lattice after being exposed to a source of ionizing radiation. In addition to the defect studies, the thermoluminescence in impurity doped alkali halides has been a subject of more investigation due to its applications in many areas such as TL dosimetry, luminescent phosphors, optical memory devices, etc.[1]. Now a day, various types of thermoluminescence dosimeters (TLDs) are available for use. The dosimetric properties of any TL material mostly depend on the kinetic parameters of its glow curve. Because it gives useful information about the mechanism of thermoluminescence in that material. Since the TL phenomenon is highly sensitive to its glow structure, it is strongly disturbed by the presence of impurities and their state of accumulation in the host lattice [2]. Over the years, there have been many studies on the thermoluminescence in impurity doped alkali halide crystals [3–5]. Earlier, the work on TL properties of some alkali halide crystals doped with trivalent rare earth has been reported from our group [6,7]. These studies were mainly concerned with the effect of impurity in host lattice, kinetic parameters, etc. on TL glow curves. The stimulated energy release these electrons may be thermal and the trapping centers involved are called electrons or holes traps. If these traps are energetically deep enough, the charge carriers may remain for a long time in the trap until they subjected to enough thermal-energy to increase their escape factor, producing electromagnetic radiation after a radiative recombination process takes place. The present paper aims at understanding the TL behaviour in KBr: Sm^{3+} Tb^{3+} crystals grown by Bridgemann Stockbarger technique.

2. Experimental Details

Single crystals of Samarium doped and terbium codoped KBr (99.99% purity) were grown using Bridgemann Stockbarger technique. Samarium was added in the form of samarium fluoride (Aldrich 99.99% purity) and terbium in the form of terbium fluoride. The crystals are grown with three different impurity concentrations 1%, 3% and 5% by weight. The results due to three concentrations were similar except high luminescence yield for crystals with a high concentration. Hence only the results pertaining to terbium and samarium concentration of 5% by weight are presented and discussed. All the measurements were performed at room temperature. TL glow curve were recorded using PC based TL analyser (integrated type TL1009I) when the crystal irradiated with γ –ray dose of 5 Gy at a heating rate of 5°C/sec. TL emission were recorded in Perkin Elmer LS 55 with excitation slit being closed. The morphologies of the samples were inspected using Scanning Electron Microscopy SEM- *JEOL-JSM 5610LV* model.

3. Thermoluminescence (TL) glow curve

Thermoluminescence spectrum gives more information about the nature of traps. Glow curve is one of the important view on investigation of any TL dosimetry material. **Fig.1** shows the TL glow curve of KBr: Sm^{3+} Tb^{3+} single crystals. Before taking the TL, the sample annealed at 400° C and then irradiated at a γ –ray dose of 5 Gy. The TL glow curve was recorded after the samples were irradiated with γ-rays for 1-hour with heating rate of 5°C/sec. TL measurements are carried out soon after irradiation of the sample to avoid the possibility of the error. Under γ-ray irradiation, many electrons transferred from valence band to the conduction band where they are free to move through crystal. Dopants play a main role in the production of TL glow curve. Trapping centre play difficult role in the photo-energy storage and thermo-stimulated phosphors. Sm^{3+} ions act as a role of creating the electron traps due to the chemically non-equivalent substitution. The exact role played by Sm^{3+} and Tb^{3+} in luminescence process could be determined by taking the TL spectrum as a function of temperature. In rare earth doped phosphors excited under gamma ray irradiation, few charge carriers (electrons or holes) may be intercepted by traps introduced in the host lattice. The captured charge carriers escape under thermal disturbances and transfer to luminescent centre, resulting in phosphorescence after the removal of source of excitation. The activation energy required for the charge carriers to escape from the traps is called the 'trap depth'. There are two types of trapping centre present in the centre of the host according to type of material doped. Shallow traps will result in a very sharp afterglow because captured charge carriers escaping too fastly whereas deep trap will cause poor afterglow because of the requirement of high activation energy which is reported in earlier work [8, 9].

The deconvolution of TL components can be roughly performed using software "Origin 5.0". This shows two well distinguished TL peaks at 448 K and 501K corresponding to two types of traps. There are various methods for evaluating the kinetic parameters from the deconvoluted TL glow curve. But we use Chen's peak shape method is used to calculate the kinetic parameters such as geometric factor μ_g, Activation energy E and Frequency factor S. According to Chen [10], there is a close connection between the kinetic order of TL process and the shape of TL band. The association between them results in a geometric factor μ_g, which equals δ/ω. The peak shape parameters τ, δ and ω were initially determined from $T_{1,}$ T_2 and T_g. $T_{1,}$ T_2 are the temperature at half intensity on either side of the peak and T_g are the glow peak temperature. τ, δ and ω are the half width on low temperature side, half width on high temperature side and full width at half maximum respectively. They are the temperature differences between $T_g - T_1$,$T_2 - T_m$ and $T_2 - T_1$. The peak shape method is generally used to calculate the order of kinetics and is obtained from the geometric factor. The geometric factor can be calculated as $\mu_g = \delta/\omega$. The order of kinetics depends on the shape of the peak. The value of μ_g is 0.42 for the first order and 0.52 for second order. In the present work, value of μ_g is 0.49 and 0.52 which shows the evidence of second order kinetics of the traps. It is clear that both the glow peaks follow second order kinetics and hence there is chance of retrapping.

The TL parameters of trap depths E, Frequency factor S and trap densities n_o can be estimated by the equation provided in the reported work [11, 12]. The loss of dosimetric information stored in the material after irradiation is strongly depends on the position of the trapping centre within the forbidden gap. The calculated trap parameters were tabulated in **table.I**. The trap densities for 448K and 501K peak were determined to be 14.99 and 24.02 $(cm^3)^{-1}$ respectively. It shows more difference and it evidence that less number of carriers trapped at shallow traps and more trapped at deep traps. The activation energy of two peaks at 448K and 501K were calculated as 1.249 eV and 1.524 eV respectively which differ slightly and it confirms that the formation of shallow and deep traps. Furthermore, the frequency factor value of 448K and 501K peaks were found to be 2.68 and 2.35 s^{-1} respectively and show minute difference. This may be because competition among various traps might be giving various escaping and retrapping probabilities. The carriers in shallow traps release too quickly, while the carriers in the deep traps are very difficult to release. So that more energy needed to detrap the electron from the deep traps. The high temperature peak at 501K is having higher trap depth and it is very useful for TLD phosphor characteristics. The characteristics of dosimeters used in TL process are related to kinetic parameters and these parameters quantitatively describe the trapping –emitting centers. Thus a dosimetric study of a TL material should be based on a good knowledge of kinetic parameters [13].

4. TL emission

Fig.2 shows TL emission spectrum KBr: Sm^{3+} Tb^{3+} single crystals at 448 K (curve a) and 501 K (curve b) respectively. **Fig.2 curve (a)** shows the emission band at 602 nm and shoulder around 528nm. **Fig.2 curve (b)** shows emission band at 589nm and shoulder around 612 nm. The emission band observed at 602nm with 448K and at 589nm with 501K is probably similar but with varied intensity. The intensity of emission is high in the emission at 501K compared to the emission at 448K. So we chose this peak for deconvolution. The deconvolution of emission spectrum at 501K made by using the software "Origin 5.0". The deconvoluted emission spectrum was shown in **Fig.3**. It shows two well separated peaks centered at 583nm and 644nm which confirms emission is only due to samarium ions. The more charge carriers were trapped in deep traps which are related to the high temperature glow peak. The trap concentration (n_o) were calculated and discussed in TL glow curve analysis. The deep trap has higher concentration of charge carriers compared to shallow traps. From table.1, n_o is higher for 501K glow peak. The higher n_o value will produce a higher initial luminescence obviously, since a large amount of traps will capture more free electrons generated by irradiation which was reported in earlier work [14]. TL emission at 583nm confirms that this emission is only due to samarium ions.

5. Scanning Electron Microscopy (SEM)

Fig.4 shows the SEM micrograph with 1000 magnification of KBr: Sm^{3+} Tb^{3+} crystals. Characterization of particles, surface morphology and size of the crystals is done mostly using SEM. The individual particle in the larger aggregated particles can be considered as a single crystallite based on their similar size. At first sight it seems that the sample is composed of particles with sizes ranging from few microns to 10 microns. From the Scanning electron micrographs of KBr: Sm^{3+} Tb^{3+} crystals, it is found that the particles are cubic in shape with sizes ranging from few microns to 10 microns and also cluster is found. The micron size of crystallite from SEM image is best suited for dosimetric applications.

6. Conclusions

Two TL peaks at 448 K and 501 K corresponding to two types of traps follow second order kinetics and hence there is chance of retrapping. The high temperature glow peak at 501K is having higher trap depth is suitable for TLD phosphor applications. Deconvoluted TL emission spectrum shows two well separated peaks centered at 583nm and 644nm which confirms emission is only due to samarium ions. From SEM image, the particles are cubic in shape with sizes ranging from few microns to 10 microns and also cluster is found. The micron size of crystallite from SEM image is best suited for dosimetric applications.

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FIGURE No.1

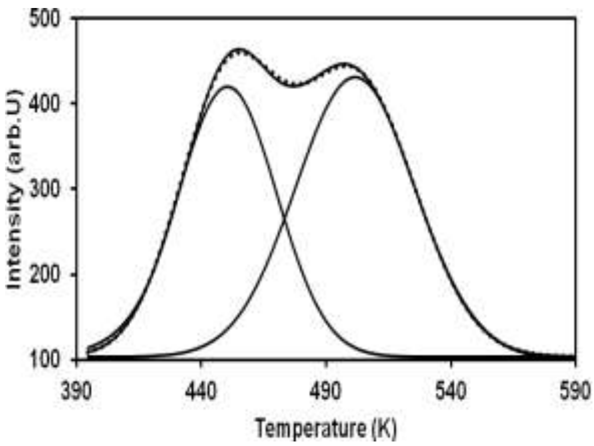

Fig.1 Deconvoluted Thermoluminescence Glow curve of KBr:Sm^{3+} Tb^{3+} crystals after γ-ray irradiation for 1-hour

FIGURE No. 2

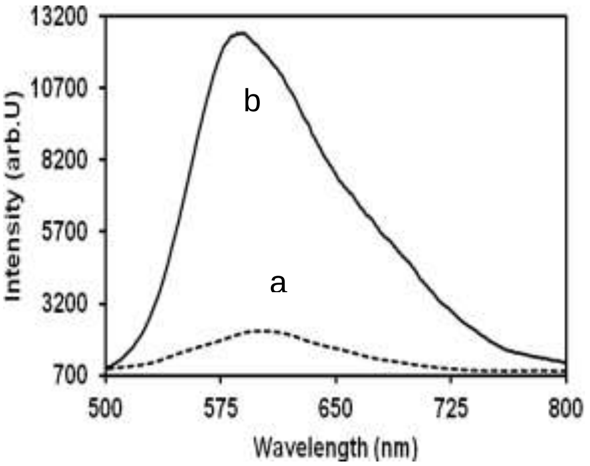

Fig.2 TL Emission Spectra of KBr :Sm^{3+} Tb^{3+} crystals at (a) 448K, (b) 501 K

FIGURE No.3

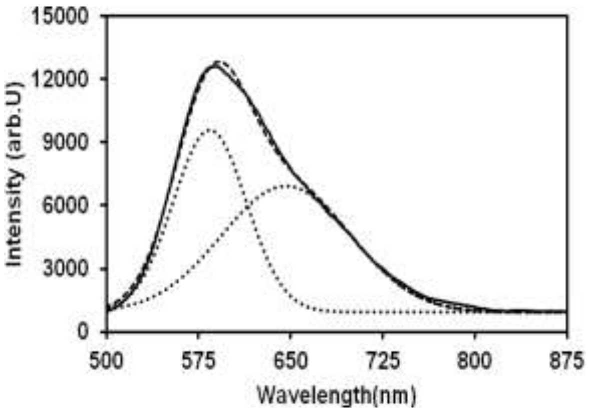

Fig.3 Deconvoluted Thermoluminescence emission spectrum of KBr: Sm^{3+} Tb^{3+} crystals at 501 K

FIGURE No.4

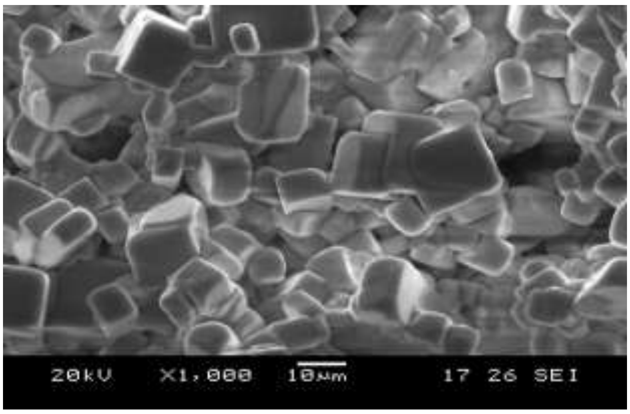

Fig.4 SEM image of KBr: Sm^{3+} Tb^{3+} crystals

TABLE No.I

Table.I Calculated Kinetic Parameters of TL glow curve of KBr : Sm^{3+} Tb^{3+} crystals

Glow Peak Temperature Tg (K)	Intensity (arb.U)	Geometric factor $\mu_g = \delta/\omega$	Activation Energy E (eV)	Frequency factor s (s^{-1})	Concentration of charge carriers n_o $(Cm^3)^{-1}$	Order of kinetics
448	417	0.52	1.249	2.68	14.99	II
501	430	0.49	1.524	2.35	24.02	II

